Copper(II) Oxide Catalyzed Ligand-Free Coupling Reaction of Heteroarenes with Bromoalkynes[1]

Biswanath Das,* Gandolla Chinna Reddy, Penagaluri Balasubramanyam, N. Salvanna

Organic Chemistry Division-1, Indian Institute of Chemical Technology, Hyderabad 500607, India
Fax +91(40)27160512; E-mail: biswanathdas@yahoo.com

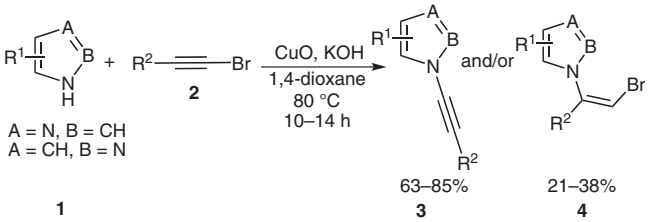
Received 26 November 2010; revised 29 November 2010

Abstract: The coupling reaction of imidazoles and pyrazoles with bromoacetylenes has efficiently been carried out under ligand-free conditions in the presence of copper(II) oxide as the catalyst using potassium hydroxide in 1,4-dioxane at 80 °C. The method is a simpler access to *N*-alk-1-ynyl- and/or *N*-(2-bromovinyl)-substituted heteroarenes.

Key words: coupling reaction, ligand-free conditions, copper(II) oxide, *N*-alkynylheteroarene, *N*-(2-bromovinyl)heteroarene

N-Alkynylheteroarenes are valuable intermediates in organic synthesis[2] as well as in medicinal chemistry.[3] They are also known to possess important biological[3,4] and photoconductive properties.[5] However, these compounds have not yet been properly explored, possibly, due to the unavailability of mild and general methods for their synthesis.[6] Earlier methods for the preparation of *N*-alkynylheteroarenes, including the elimination of haloenamines,[7a] coupling of alkynyliodonium salts,[7b] and isomerization of the propargyl group,[7c] require several steps. These compounds have also been prepared using ligand-assisted copper(I) iodide as the catalyst and microwave irradiation.[8] Herein, we report a ligand-free efficient synthesis of *N*-alkynylheteroarenes.

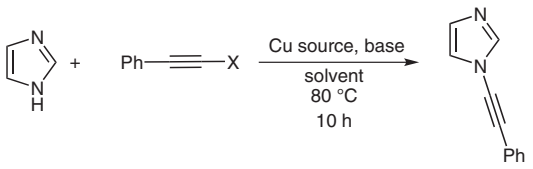
In continuation of our work[9] on the development of useful synthetic methodologies, we have discovered that the treatment of heteroarenes with bromoalkynes in the presence of a catalytic amount of copper(II) oxide with potassium hydroxide as the base in 1,4-dioxane afforded the corresponding *N*-alkynyl- and/or *N*-(2-bromovinyl)-substituted heteroarenes at 80 °C (Scheme 1).



Scheme 1

Initially the reaction of imidazole with bromo- and iodo(phenyl)acetylenes was studied under ligand-free conditions using various copper sources and different bases as well as solvents (Table 1). Bromo(phenyl)acetylene was found to undergo C–N coupling reaction with imidazole to form 1-(phenylethynyl)-1*H*-imidazole (entries 1–10) while iodo(phenyl)acetylene was almost inactive (entries 11 and 12). The reaction was carried out at 80 °C; no product was isolated at room temperature. Various copper catalysts were examined for the reaction of bromo(phenyl)acetylene with imidazole, copper(II) oxide gave the best yields (cf. entry 5 vs. entries 4, 8, and 10). It is noteworthy that copper(I) iodide was used as the catalyst in earlier cases of the reaction of imidazoles with bromoalkynes,[8] however, under the present conditions its

Table 1 Optimization of Reaction Conditions[a]



Entry	X	Cu Source	Solvent	Base	Yield[b] (%)
1	Br	CuI	toluene	KOH	50
2	Br	CuI	1,4-dioxane	Cs_2CO_3	45
3	Br	CuI	1,4-dioxane	K_3PO_4	48
4	Br	$Cu(OTf)_2$	1,4-dioxane	K_3PO_4	22
5	Br	CuO	1,4-dioxane	KOH	80
6	Br	CuO	toluene	KOH	68
7	Br	CuO	1,4-dioxane	Cs_2CO_3	72
8	Br	$Cu(OAc)_2$	1,4-dioxane	KOH	25
9	Br	$Cu(OTf)_2$	1,4-dioxane	KOH	trace
10	Br	$CuSO_4 \cdot 5\,H_2O$	1,4-dioxane	KOH	30
11	I	CuI	toluene	Cs_2CO_3	–[c]
12	I	CuO	1,4-dioxane	KOH	–[c]

[a] Reaction conditions: imidazole (1.0 mmol), haloalkyne (1.0 mmol), copper source (2.0 mol%), base (2.0 equiv), solvent (3 mL), 80 °C, 10 h.
[b] Isolated yield.
[c] No reaction.

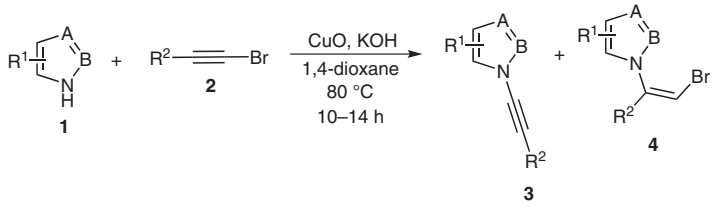
SYNTHESIS 2011, No. 5, pp 0816–0820
Advanced online publication: 11.01.2011
DOI: 10.1055/s-0030-1258403; Art ID: Z29810SS

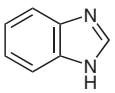
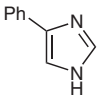
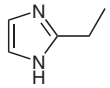
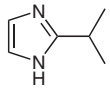
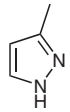
activity was not impressive (entries 1–3). Next examining the solvent showed that 1,4-dioxane gave superior yields to toluene (entry 5 vs. entry 6). Finally potassium hydroxide and cesium carbonate were examined as the base, potassium hydroxide gave a better yield (80% vs. 72%, entry 5 vs. entry 7). The optimal conditions were using the bromoacetylene as substrate with copper(II) oxide as the catalyst, potassium hydroxide as the base, and 1,4-dioxane as the solvent at 80 °C.

A series of *N*-alkynylimidazoles **3** were prepared (Table 2) using the optimized conditions (Scheme 1). Various substituted imidazoles, including benzimidazoles, were employed (entries 1–13). Both aromatic and aliphatic bromoacetylenes underwent the conversion smoothly. Even bromo(6-methoxy-2-naphthyl)acetylene afforded the corresponding products (entries 3, 6, 9, and 16). The conversion was complete within 10–14 hours and the *N*-alkynylimidazoles **3** were formed in high yields (67–85%).

When 4-phenyl-1*H*-imidazole was treated with bromo(phenyl)acetylene, a mixture of 1,4- and 1,5-regioisomers of **3g** were produced in the ratio of 3:2 (entry 7). However, reaction of 4-phenyl-1*H*-imidazole with bromo(4-pentylphenyl)acetylene (entry 8) or with bromo(6-methoxy-2-naphthyl)acetylene (entry 9) gave only the 1,4-regioisomers. The reaction of 2-ethyl-1*H*-imidazole with bromo(phenyl)acetylene under these conditions yielded 2-ethyl-1-(phenylethynyl)-1*H*-imidazole (**3l**) as the major product (71%) together with the minor product 1-(2-bromo-1-phenylvinyl)-2-ethyl-1*H*-imidazole (**4l**) (22%) (entry 12). A similar result was also observed when benzimidazole was treated with bromo(6-methoxy-2-

Table 2 C–N Coupling of Bromoalkynes with Imidazoles and Pyrazoles[a,b]



Entry	Imidazole/Pyrazole **1**	R^2	Time (h)	Product	Yield[c] (%) of **3**	Yield[c] (%) of **4**
1		Ph	10	**3a**	80	–
2		4-Me$(CH_2)_4C_6H_4$	12	**3b**	78	–
3		6-methoxy-2-naphthyl	14	**3c**	73	–
4		Ph	11	**3d**	76	–
5		4-Me$(CH_2)_4C_6H_4$	12	**4e**	–	38
6		6-methoxy-2-naphthyl	13	**3f/4f**	67	24
7		Ph	10	**3g**	81[d]	–
8		4-Me$(CH_2)_4C_6H_4$	13	**3h**	82	–
9		6-methoxy-2-naphthyl	13	**3i**	85	–
10		$(CH_2)_5Me$	14	**3j**	78	–
11		Ph	10	**3k**	68	–
12		Ph	12	**3l/4l**	71	22
13		Ph	13	**3m**	79	–
14		Ph	10	**3n**	69	–
15		4-Me$(CH_2)_4C_6H_4$	11	**3o/4o**	63	21
16		6-methoxy-2-naphthyl	14	**3p**	72	–
17		Ph	12	**3q**	69	–

[a] Reaction conditions: imidazole/pyrazole (1.0 mmol), haloalkyne (1.0 mmol), CuO (2.0 mol%), KOH (2.0 equiv), 1,4-dioxane (3 mL), 80 °C.
[b] Compounds **3a**, **3d**, **3g**, **3j, 3k**, **3n** are known.[8]
[c] Isolated yield.
[d] Ratio of 1,4- to 1,5-regioisomeric products was (3:2).

naphthyl)acetylene (entry 6). However, when benzimidazole was treated with bromo(4-pentylphenyl)acetylene, 1-[2-bromo-1-(4-pentylphenyl)vinyl]-1*H*-benzimidazole (**4e**) was formed as the sole product, though its yield was low (38%) (entry 5). The olefinic double bond in all the *N*-(2-bromovinyl) products was in the *Z*-configuration as the olefinic proton did not show any NOESY correlation with the protons of the imidazole core.

The above reaction was also successfully applied to the C–N coupling of bromoalkynes with pyrazoles (Scheme 1) to produce a series of *N*-alkynylpyrazoles (entries 14–17); the products were formed in good yields (63–72%) within 10–14 h. The reaction of pyrazole with bromo(4-pentylphenyl)acetylene afforded both *N*-alkynylpyrazole **3o** (major, 63%) and *N*-(2-bromovinyl)pyrazole **4o** (minor, 21%) (entry 15). The structures of all the products were established from their spectral (IR, ^{1}H and ^{13}C NMR, ESIMS and HRESIMS) data.

In conclusion, we have developed a simple and efficient coupling reaction of heteroarenes with bromoacetylenes using copper(II) oxide (as a catalyst) and potassium hydroxide in 1,4-dioxane to prepare *N*-alk-1-ynyl- and/or *N*-(2-bromovinyl)-substituted heteroarenes under ligand-free conditions. Fourteen new compounds have been prepared.

TLC used silica gel F_{254} plates; the spots were examined under UV light and then developed by I_2 vapor. Column chromatography was performed with silica gel (BDH 100–200 mesh). Solvents were purified according to standard procedures. The spectra were recorded with the following instruments; IR: Perkin-Elmer RX FT-IR spectrophotometer; NMR: Varian Gemini 200 MHz (^{1}H) and 50 MHz (^{13}C) spectrometer; ESIMS: VG-Autospec micromass. Organic extracts were dried over anhyd Na_2SO_4.

N-Alkynylation of Imidazoles and Pyrazoles; General Procedure

To a soln of bromoacetylene **2** (1.0 mmol) in anhyd 1,4-dioxane (3 mL) were added imidazole or pyrazole **1** (1.0 mmol), KOH (2.0 equiv), and CuO (2.0 mol%). The mixture was heated in an oil bath at 80 °C for 10–14 h. The reaction was monitored by TLC. Upon completion, the mixture was cooled to r.t., the solvent was evaporated, and the mixture was diluted with cold H_2O (10 mL). This was extracted with EtOAc (2 × 10 mL). The combined organic layers were dried (anhyd Na_2SO_4), concentrated in vacuo, and purified by column chromatography (silica gel, Merck, 60–120 mesh, 1–4% EtOAc–hexane) to afford pure product(s).

1-[(4-Pentylphenyl)ethynyl]-1*H*-imidazole (3b)

IR (KBr): 2264, 1677, 1482, 130 cm^{-1}.

^{1}H NMR (200 MHz, $CDCl_3$): δ = 7.79 (s, 1 H), 7.39 (d, *J* = 8.0 Hz, 2 H), 7.18 (s, 1 H), 7.12 (d, *J* = 8.0 Hz, 2 H), 7.05 (s, 1 H), 2.60 (t, *J* = 7.0 Hz, 2 H), 1.70–1.51 (m, 3 H), 1.38–1.29 (m, 3 H), 0.90 (t, *J* = 7.0 Hz, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 144.2, 140.0, 132.1, 129.3, 129.0, 122.1, 118.5, 77.9, 70.8, 36.1, 31.7, 31.2, 22.8, 14.1.

MS (ESI): *m*/*z* = 239 [M + H]$^+$.

HRMS (ESI): *m*/*z* [M + H]$^+$ calcd for $C_{16}H_{19}N_2$: 239.1548; found: 239.1559.

1-[(6-Methoxy-2-naphthyl)ethynyl]-1*H*-imidazole (3c)

IR (KBr): 2309, 1638, 1498, 1389, 1216 cm^{-1}.

^{1}H NMR (200 MHz, $CDCl_3$): δ = 7.98 (s, 1 H), 7.88 (s, 1 H), 7.72 (d, *J* = 8.0 Hz, 2 H), 7.49 (d, *J* = 8.0 Hz, 1 H), 7.28–7.05 (m, 4 H), 3.92 (s, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 159.2, 132.0, 129.7, 129.5, 128.9, 127.3, 122.1, 120.0, 106.1, 78.8, 69.0, 55.4.

MS (ESI): *m*/*z* = 249 [M + H]$^+$.

HRMS (ESI): *m*/*z* [M + H]$^+$ calcd for $C_{16}H_{13}N_2O$: 249.1027; found: 249.1019.

1-[2-Bromo-1-(4-pentylphenyl)vinyl]-1*H*-benzimidazole (4e)

IR (KBr): 2310, 1644, 1456 cm^{-1}.

^{1}H NMR (200 MHz, $CDCl_3$): δ = 8.01 (m, 1 H), 7.82 (m, 1 H), 7.36–7.01 (m, 7 H), 6.92 (s, 1 H), 2.59 (t, *J* = 7.0 Hz, 2 H), 1.71–1.52 (m, 2 H), 1.40–1.24 (m, 4 H), 0.88 (t, *J* = 7.0 Hz, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 146.6, 141.0, 139.0, 135.5, 129.6, 129.0, 126.1, 124.1, 122.9, 120.6, 111.8, 103.7, 36.0, 31.8, 31.1, 22.8, 14.2.

MS (ESI): *m*/*z* = 369, 371 [M + H]$^+$.

HRMS (ESI): *m*/*z* [M + H]$^+$ calcd for $C_{20}H_{22}BrN_2$: 369.0966; found: 369.0960.

1-[(6-Methoxy-2-naphthyl)ethynyl]-1*H*-benzimidazole (3f)

IR (KBr): 2265, 1617, 1528, 1489, 1245 cm^{-1}.

^{1}H NMR (200 MHz, $CDCl_3$): δ = 8.20 (s, 1 H), 8.02 (s, 1 H), 7.88 (d, *J* = 8.0 Hz, 1 H), 7.80–7.66 (m, 3 H), 7.58 (d, *J* = 8.0 Hz, 1 H), 7.50–7.33 (m, 2 H), 7.28–7.12 (m, 2 H), 3.94 (s, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 159.0, 144.0, 132.2, 129.5, 129.2, 128.6, 128.5, 127.3, 127.2, 125.1, 124.4, 121.1, 120.0, 116.2, 110.9, 105.8, 77.8, 74.2, 55.3.

MS (ESI): *m*/*z* = 299 [M + H]$^+$.

HRMS (ESI): *m*/*z* [M + H]$^+$ calcd for $C_{20}H_{15}N_2O$: 299.1184; found: 299.1170.

1-[2-Bromo-1-(6-methoxy-2-naphthyl)vinyl]-1*H*-benzimidazole (4f)

IR (KBr): 2267, 1620, 1505, 1272 cm^{-1}.

^{1}H NMR (200 MHz, $CDCl_3$): δ = 8.19 (s, 1 H), 7.91 (d, *J* = 8.0 Hz, 1 H), 7.74–7.61 (m, 2 H), 7.52 (s, 1 H), 7.40–7.01 (m, 7 H), 3.91 (s, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 159.4, 135.1, 130.2, 128.7, 128.2, 126.2, 124.0, 123.8, 123.2, 120.2, 120.0, 111.9, 106.0, 104.1, 55.2.

MS (ESI): *m*/*z* = 379, 381 [M + H]$^+$.

HRMS (ESI): *m*/*z* [M + H]$^+$ calcd for $C_{20}H_{16}BrN_2O$: 379.0445; found: 379.0435.

1-[(4-Pentylphenyl)ethynyl]-4-phenyl-1*H*-imidazole (3h)

IR (KBr): 2250, 1605, 1485, 1413 cm^{-1}.

^{1}H NMR (200 MHz, $CDCl_3$): δ = 7.81–7.70 (m, 3 H), 7.42–7.30 (m, 5 H), 7.24 (t, *J* = 8.0 Hz, 1 H), 7.12 (d, *J* = 8.0 Hz, 2 H), 2.62 (t, *J* = 7.0 Hz, 2 H), 1.70–1.52 (m, 3 H), 1.39–1.22 (m, 3 H), 0.89 (t, *J* = 7.0 Hz, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 144.2, 142.0, 140.1, 133.0, 131.9, 128.9, 127.7, 125.6, 118.2, 117.0, 77.7, 71.0, 36.1, 31.2, 30.8, 22.5, 13.8.

MS (ESI): *m*/*z* = 315 [M + H]$^+$.

HRMS (ESI): *m*/*z* [M + H]$^+$ calcd for $C_{22}H_{23}N_2$: 315.1861; found: 315.1854.

1-[(6-Methoxy-2-naphthyl)ethynyl]-4-phenyl-1*H*-imidazole (3i)
IR (KBr): 2254, 1622, 1493, 1383, 1263 cm^{-1}.

^1H NMR (200 MHz, $CDCl_3$): δ = 7.99 (s, 1 H), 7.90 (s, 1 H), 7.82 (d, *J* = 8.0 Hz, 2 H), 7.73 (d, *J* = 8.0 Hz, 2 H), 7.52–7.35 (m, 4 H), 7.31–7.10 (m, 3 H), 3.92 (s, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 159.0, 140.1, 131.9, 129.6, 129.0, 127.9, 127.1, 125.1, 119.9, 119.8, 117.2, 117.0, 105.9, 78.0, 71.2, 55.5.

MS (ESI): *m/z* = 325 $[M + H]^+$.

HRMS (ESI): *m/z* $[M + H]^+$ calcd for $C_{22}H_{17}N_2O$: 325.1340; found: 325.1326.

2-Ethyl-1-(phenylethynyl)-1*H*-imidazole (3l)
IR (KBr): 2264, 1681, 1503, 1428 cm^{-1}.

^1H NMR (200 MHz, $CDCl_3$): δ = 7.56–7.47 (m, 2 H), 7.43–7.32 (m, 3 H), 7.12 (s, 1 H), 6.93 (s, 1 H), 2.91 (q, *J* = 7.0 Hz, 2 H), 1.40 (t, *J* = 7.0 Hz, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 146.9, 131.8, 129.0, 128.8, 127.9, 121.5, 78.0, 73.4, 20.7, 11.3.

MS (ESI): *m/z* = 197 $[M + H]^+$.

HRMS (ESI): *m/z* $[M + H]^+$ calcd for $C_{13}H_{13}N_2$: 197.1078; found: 197.1086.

1-(2-Bromo-1-phenylvinyl)-2-ethyl-1*H*-imidazole (4l)
IR (KBr): 2309, 1648, 1435, 1281 cm^{-1}.

^1H NMR (200 MHz, $CDCl_3$): δ = 7.40–7.31 (m, 3 H), 7.18–7.10 (m, 3 H), 7.07 (s, 1 H), 6.89 (s, 1 H), 2.48 (q, *J* = 7.0 Hz, 2 H), 1.21 (t, *J* = 7.0 Hz, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 150.0, 141.5, 131.1, 130.0, 129.2, 127.9, 125.2, 119.8, 116.9, 20.2, 11.7.

MS (ESI): *m/z* = 277, 279 $[M + H]^+$.

HRMS (ESI): *m/z* $[M + H]^+$ calcd for $C_{13}H_{14}BrN_2$: 277.0340; found: 277.0351.

2-Isopropyl-1-(phenylethynyl)-1*H*-imidazole (3m)
IR (KBr): 2263, 1500, 1426, 1253 cm^{-1}.

^1H NMR (200 MHz, $CDCl_3$): δ = 7.52–7.45 (m, 2 H), 7.40–7.32 (m, 3 H), 7.10 (s, 1 H), 6.91 (s, 1 H), 3.32 (m, 1 H), 1.41 (d, *J* = 7.0 Hz, 6 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 157.0, 131.8, 129.1, 128.9, 127.9, 121.1, 78.1, 72.6, 27.0, 20.9.

MS (ESI): *m/z* = 211 $[M + H]^+$.

HRMS (ESI): *m/z* $[M + H]^+$ calcd for $C_{14}H_{15}N_2$: 211.1235; found: 211.1233.

1-[(4-Pentylphenyl)ethynyl]-1*H*-pyrazole (3o)
IR (KBr): 2257, 1609, 1434, 1397, 1261 cm^{-1}.

^1H NMR (200 MHz, $CDCl_3$): δ = 7.68 (m, 1 H), 7.60 (m, 1 H), 7.35 (d, *J* = 8.0 Hz, 2 H), 7.09 (d, *J* = 8.0 Hz, 2 H), 6.29 (m, 1 H), 2.52 (t, *J* = 7.0 Hz, 2 H), 1.62–1.48 (m, 3 H), 1.33–1.14 (m, 3 H), 0.82 (t, *J* = 7.0 Hz, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 144.1, 142.2, 134.2, 131.8, 128.9, 118.2, 107.2, 77.3, 67.3, 36.0, 31.6, 31.0, 22.1, 13.5.

MS (ESI): *m/z* = 239 $[M + H]^+$.

HRMS (ESI): *m/z* $[M + H]^+$ calcd for $C_{16}H_{19}N_2$: 239.1548; found: 239.1552.

1-[2-Bromo-1-(4-pentylphenyl)vinyl]-1*H*-pyrazole (4o)
IR (KBr): 2309, 1652, 1616, 1512 cm^{-1}.

^1H NMR (200 MHz, $CDCl_3$): δ = 7.74 (m, 1 H), 7.67 (m, 1 H), 7.12 (d, *J* = 8.0 Hz, 2 H), 7.01 (d, *J* = 8.0 Hz, 2 H), 6.62 (s, 1 H), 6.42 (m, 1 H), 2.01 (t, *J* = 7.0 Hz, 2 H), 1.69–1.52 (m, 2 H), 1.39–1.21 (m, 4 H), 0.90 (t, *J* = 7.0 Hz, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 145.0, 141.1, 133.4, 131.8, 128.9, 128.8, 126.3, 106.2, 100.6, 35.8, 31.2, 30.7, 22.3, 14.0.

MS (ESI): *m/z* = 319, 321 $[M + H]^+$.

HRMS (ESI): *m/z* $[M + H]^+$ calcd for $C_{16}H_{20}BrN_2$: 319.0809; found: 319.0810.

1-[(6-Methoxy-2-naphthyl)ethynyl]-1*H*-pyrazole (3p)
IR (KBr): 2235, 1628, 1458, 1388, 1261 cm^{-1}.

^1H NMR (200 MHz, $CDCl_3$): δ = 8.01 (s, 1 H), 7.83–7.69 (m, 4 H), 7.52 (d, *J* = 8.0 Hz, 1 H), 7.21–7.09 (m, 2 H), 6.40 (m, 1 H), 3.91 (s, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 155.7, 143.9, 139.0, 131.9, 129.3, 129.1, 127.1, 119.2, 106.0, 76.2, 68.9, 55.7.

MS (ESI): *m/z* = 249 $[M + H]^+$.

HRMS (ESI): *m/z* $[M + H]^+$ calcd for $C_{16}H_{13}N_2O$: 249.1027; found: 249.1028.

3-Methyl-1-(phenylethynyl)-1*H*-pyrazole (3q)
IR (KBr): 2263, 1558, 1454, 1411, 1281 cm^{-1}.

^1H NMR (200 MHz, $CDCl_3$): δ = 7.54–7.48 (m, 3 H), 7.39–7.30 (m, 3 H), 6.06 (m, 1 H), 2.49 (s, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 142.7, 131.5, 130.9, 128.8, 128.7, 128.6, 105.6, 79.9, 71.9, 11.3.

MS (ESI): *m/z* = 183 $[M + H]^+$.

HRMS (ESI): *m/z* $[M + H]^+$ calcd for $C_{12}H_{11}N_2$: 183.0922; found: 183.0918.

Acknowledgment

The author thanks CSIR and UGC, New Delhi for financial assistance.

References

(1) Part 217 in the series 'Studies on novel synthetic methodologies'.
(2) Ishihara, T.; Mantani, T.; Konno, T.; Yamanaka, H. *Tetrahedron* **2006**, *62*, 3783.
(3) Joshi, R. V.; Xu, Z. Q.; Ksebati, M. B.; Kessel, D.; Corbett, T. H.; Drach, J. C.; Zemlicka, J. *J. Chem. Soc., Perkin Trans. 1* **1994**, 1089.
(4) (a) Reisch, J.; Seeger, U. *Arch. Pharm. (Weinheim, Ger.)* **1977**, *310*, 851. (b) Parsons, C. G. R.; Jirgensons, A.; Jaunzeme, I.; Kalvinsh, I.; Henrich, M.; Vanejevs, M.; Weil, T.; Kauss, V.; Danysz, W.; Jatzke, C. WO 2007023290, **2007**.
(5) Brabec, C.; Scharber, M.; Johansson, H.; Comoretto, D.; Dellepiane, G.; Moggio, I.; Cravino, A.; Hummelen, J. C.; Sariciftci, N. S. *Synth. Met.* **1999**, *101*, 298.
(6) (a) Mulder, J. A.; Kurtz, K. C. M.; Hsung, R. P. *Synlett* **2003**, 1379. (b) Katritzky, A. R.; Jiang, R.; Singh, S. K. *Heterocycles* **2004**, *63*, 1455.
(7) (a) Brandsma, L.; Mal'kina, A. G.; Trofimov, B. A. *Synth. Commun.* **1994**, *24*, 2721. (b) Kitamura, T.; Tashi, N.; Tsuda, K.; Chen, H.; Fujiwara, T. *Heterocycles* **2000**, *52*, 303. (c) Wei, L.; Mulder, J. A.; Xiong, H.; Zificsak, C. A.; Douglas, C. J.; Hsung, R. P. *Tetrahedron* **2001**, *57*, 459.

(8) (a) Laroche, C.; Li, J.; Freyer, M. W.; Kerwin, S. M. *J. Org. Chem.* **2008**, *73*, 6462. (b) Burley, G. A.; Davies, D. L.; Griffith, G. A.; Lee, M.; Sing, K. *J. Org. Chem.* **2010**, *75*, 980.

(9) (a) Das, B.; Damodar, K.; Bhunia, N. *J. Org. Chem.* **2009**, *74*, 5607. (b) Das, B.; Satyalakshmi, G.; Suneel, K.; Damodar, K. *J. Org. Chem.* **2009**, *74*, 8400. (c) Das, B.; Reddy, Ch. R.; Kumar, D. N.; Krishnaiah, M.; Narendar, R. *Synlett* **2010**, 391.