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The Use of a Bifunctional Copper Catalyst in the Cross-Coupling Reactions of Aryl and Heteroaryl Halides with Terminal Alkynes

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Copper-catalyzed cross-coupling reactions of aryl halides and heteroaryl halides with terminal alkynes were carried out in DMF at 110–130 °C to provide the corresponding coupling products in satisfactory-to-good yields by using inexpensive 8-hydoxyquinoline as the ligand. Noteworthy is that the bifunctional copper catalyst was successfully applied in this alkynylation reaction for the first time.

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Introduction

The Sonogashira-type cross coupling reaction of aryl halides with terminal alkynes[1] has gained great attention in organic synthesis, because it has been applied widely in the synthesis of molecules containing an acetylenic moiety, which is important for various functional organic materials as well as biologically important molecules and their intermediates.[2] Actually, typical Sonogashira coupling is the alkynylation of aryl halides in the presence of $PdCl_2(PPh_3)_2$, $PdCl_2/PPh_3$, or $Pd(PPh_3)_4$ together with CuI as the cocatalyst and large amounts of amines as the solvents or cosolvents.[3] Recently, such coupling reactions were modified by using different catalytic systems in the absence of copper salts,[4] including palladium(0)[5] and gold(III),[6] but they were both limited to industrial use owing to the expensive cost of palladium and gold. In addition, nickel-complex systems are also not economic in this coupling reaction, and the scope of the substrates is only limited to aryl iodides together with an additional metal catalyst.[7] Recently, Chang reported that the copper-free alkynylation of aryl iodides was investigated in the presence of a new catalyst system of ruthenium supported on alumina.[8]

Thus, copper(I) alone as a metal catalyst is more attractive, which is considered as the better choice for the coupling of aryl halides and terminal alkynes,[9] because copper compounds are cheap and the system is simple and mild. Certainly, the ligand is very important to the reaction. Recent studies show that a copper catalyst together with a phosphorus compound (such as triphenylphosphane[10]), bis(nitrogen) compounds {such as **1**, 10-phenanthroline,[11] ethylenediamine,[12] DABCO (1,4-diazabicyclo[2.2.2]-octane),[13] and *N*,*N*-dimethylglycine}[14] are effective catalytic systems in this alkynylation of aryl halides. Differently, our recent results showed that *rac*-BINOL (1,1′-binaphthyl-2,2′-diol) as an O,O ligand is also effective for this coupling reaction,[15] but a simple N,O ligand has not been reported in the copper-catalyzed alkynylation reaction until now.

With an interest in asymmetric catalysis,[16] we find that bifunctional catalysts have been tremendously developed for various asymmetric transition processes.[17] Indeed, these catalysts represent bifunctional catalytic systems, where different reactive sites account for the activation of both substrates. However, as far as we know, until now there has been no report on the bifunctional catalyst system for the racemic version of these reactions, such as cross-coupling reactions. On the basis of the mechanistic consideration of copper-catalyzed coupling reactions, we expected that possible bifunctional catalyst system **1** generated in situ (Figure 1),[18] where two different parts could attach both the electrophilic and nucleophilic substrates, could lead to double activation and would be an efficient catalyst for the alkynylation reaction.

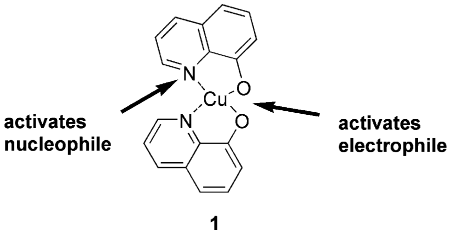

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Figure 1. The possible copper-based bifunctional catalyst system.

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Results and Discussion

Thus, the catalytic activity of CuI/8-hydoxyquinoline (**2**) was firstly tested in the coupling reaction of 1-iodobenzene with phenylacetylene, and the coupling product was obtained in 83% yield (Figure 2). It suggested that the bifunctional catalyst showed good catalytic efficiency. For the purpose of comparison, other commonly used ligands, including TMDA (*N*,*N*,*N*′,*N*′-tetramethylethane-1,2-diamine; **3**), DBU (1,8-diazabicyclo[5.4.0]undec-7-ene; **4**), DABCO (**5**), hexamethylenetetramine (**6**), 2,2′-bipyridine (**7**), 1,10-phenanthroline (**8**),[19] 2,9-dimethyl-1,10-phenanthroline (**9**),[19] L-proline (**10**), and *rac*-BINOL (**11**), have also been investigated in the coupling reaction under the same conditions. From Figure 2, it can be seen that **2** gave an identical result to that of **5**, which was reported by Li.[12]

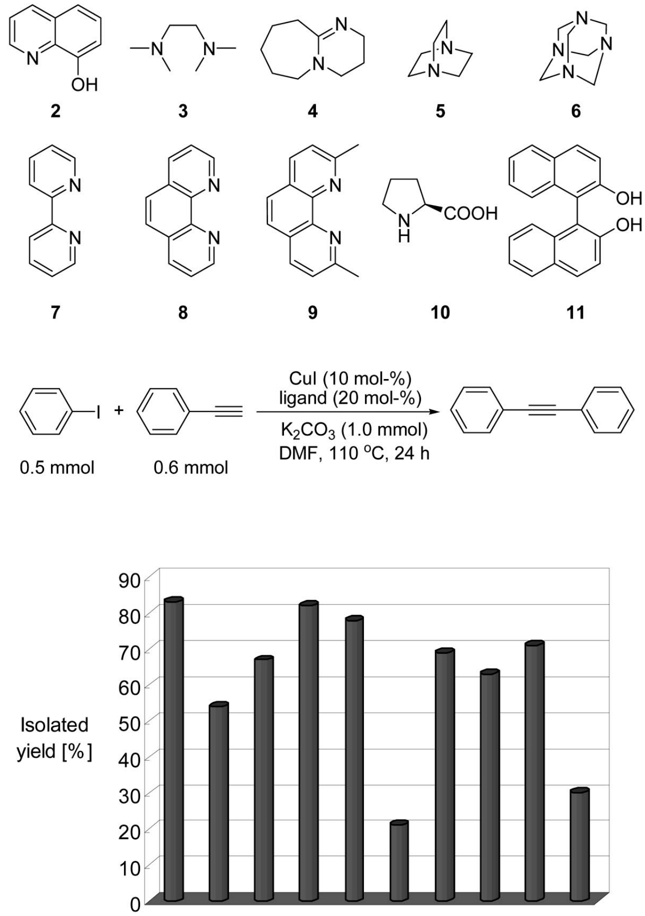
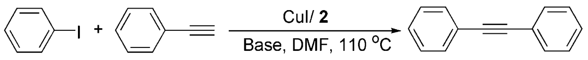


Figure 2. Various ligands investigated in the coupling of iodobenzene and phenylacetylene.

Therefore, **2** was chosen as the model ligand for screening the catalytic conditions in the coupling reaction, and the results are listed in Table 1. Firstly, a series of bases, including inorganic and organic bases (such as Cs_2CO_3, KOH, K_3PO_4, NaOAc, K_2CO_3, and Et_3N) were then screened, and Cs_2CO_3 as the base gave the best result (92%; Table 1, Entry 6). Different solvents including dimethyl sulfoxide (DMSO), 1,4-dioxane, and toluene were also investigated, and DMF gave the highest yield of the desired product. A control experiment showed that CuI was necessary for the reaction, as no product was obtained in its absence (Table 1, Entry 8). Without ligand **2**, the yield of the corresponding cross-coupling product was low (24%; Table 1, Entry 9). When the catalytic reaction was performed directly in air, the yield was unsatisfactory, and actually, more self-coupling of the alkyne was obtained. This could partly be due to the presence of O_2 from the air, which improves the homocoupling reaction as reported.[20]

Table 1. The coupling of iodobenzene and phenylacetylene catalyzed by CuI/**2**.[a]

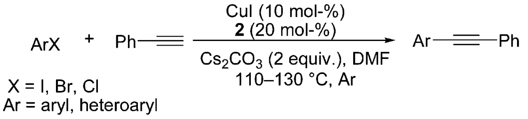


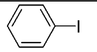
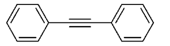
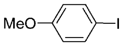
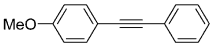
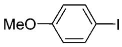
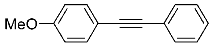
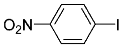
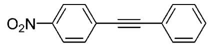
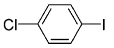
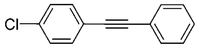
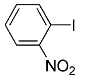
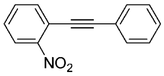
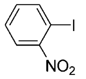
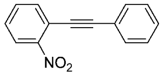
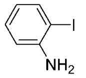
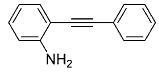
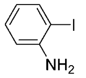
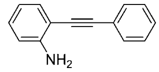
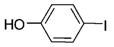
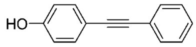
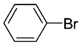
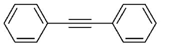
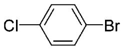
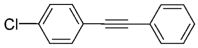
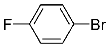
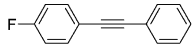
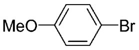
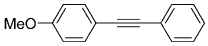
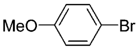
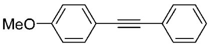
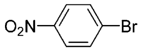
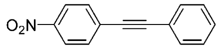
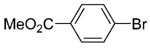
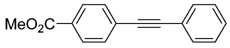
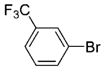
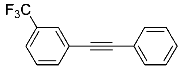
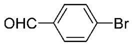
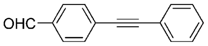
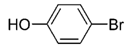
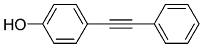
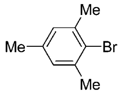
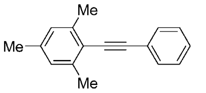
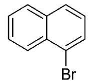
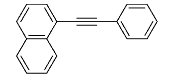
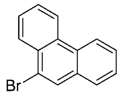
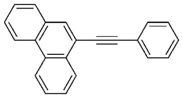
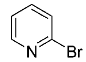
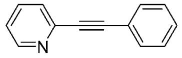
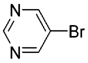
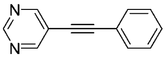
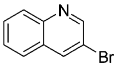
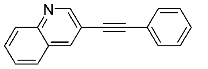
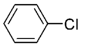
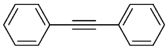
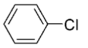
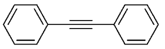
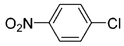
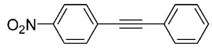
Entry	Ligand	Base	Time [h]	Yield [%][b]
1	**2**	K_2CO_3	24	83
2	**2**	K_3PO_4	24	87
3	**2**	NaOAc	24	48
4	**2**	KF	24	19
5	**2**	KOH	24	54
6	**2**	Cs_2CO_3	24	92
7	**2**	Et_3N	24	trace
8[c]	**2**	Cs_2CO_3	24	no reaction
9	–	Cs_2CO_3	24	24
10[d]	**2**	Cs_2CO_3	24	55

[a] Reaction conditions: under an argon atmosphere, iodobenzene (0.5 mmol), phenylacetylene (0.6 mmol), CuI (0.05 mmol), ligand (0.1 mmol), base (1.0 mmol), DMF (2 mL), 110 °C. [b] Isolated yield (based on iodobenzene). [c] No CuI. [d] The catalytic reaction was performed in air.

With these optimized reaction conditions in hand, we then turned our interest to the reaction generality. The results are summarized in Table 2 and Schemes 2–4. As shown in Table 2, the combination of CuI and **2** was an effective catalytic system for the alkynylation coupling reactions of various aryl halides. In the presence of CuI (10 mol-%), **2** (20 mol-%), and Cs_2CO_3 (2 equiv.), most of the reactions proceeded smoothly to afford the corresponding enynes in good-to-excellent yields. As for the *para*-substituted aryl iodides, the electronic nature of the substituents has little influence on the efficiency of the couplings (Table 2, Entries 1–5), although the reaction was slightly retarded with substrates bearing electron-donating groups (Table 2, Entry 2). To our delight, an elevated temperature and a prolonged reaction time were helpful to obtain satisfactory results (Table 2, Entry 3). For the sterically hindered *ortho*-substituted aryl iodides, the yields of the desired products were moderate (Table 2, Entries 6–9). Formation of the cyclic product was not observed in the coupling of 2-iodoaniline with phenylacetylene (Table 2, Entries 8 and 9). When 4-iodophenol was used, the corresponding coupling product was obtained in moderate yield (68%; Table 2, Entry 10). When the substrates were aryl bromides, a higher reaction temperature and a prolonged reaction time were required to get satisfactory results (Table 2, Entries 11–23). Furthermore, the coupling of heteroaryl bromides with phenylacetylene afforded good results (Table 2, Entries 24

Table 2. The cross coupling of aryl halides or heteroaryl halides and phenylacetylene catalyzed by CuI/**2**.[a]

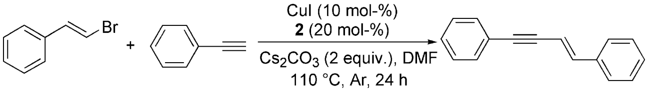


Entry	ArX	Product	T [°C]	t [h]	Yield [%][b]
1			110	24	92
2			110	24	86
3			110	30	95
4			110	24	90
5			110	24	90
6			110	24	60
7			130	30	64
8			110	24	62
9			130	30	78
10			130	24	68
11			110	24	85
12			130	30	67
13			130	22	89
14			110	24	70
15			130	30	78
16			110	24	71
17			130	24	54
18			130	36	28
19			130	30	61
20			130	24	20
21			130	24	20
22			130	24	78
23			130	24	25
24			130	24	94
25			130	24	96
26			130	24	23
27			110	20	10
28[c]			130	30	38
29[c]			130	30	34

[a] Reaction conditions: under an argon atmosphere, iodobenzene (0.5 mmol), phenylacetylene (0.6 mmol), CuI (0.05 mmol), **2** (0.1 mmol), Cs_2CO_3 (1.0 mmol), DMF (2 mL), 110–130 °C. [b] Isolated yield (based on aryl halide). [c] TBAB (1.2 equiv.) was used as the additive.

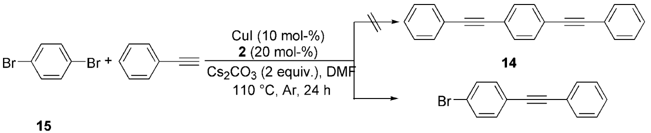
and 25). However, a poor result was acquired when 3-bromoquinoline was employed as the heteroaryl halide (Table 2, Entry 26). Interestingly, the coupling of the aryl chlorides and phenylacetylene proceeded smoothly when TBAB (tetrabutylammonium bromide; 1.2 equiv.) was used as the additive (Table 2, Entries 27–29).[12]

Because of the importance of the 1,3-enyne unit in naturally occurring and biologically active compounds,[21] many methods have been developed to create this unit.[22] Thus, as demonstrated in Scheme 1, the coupling of (*E*)-β-bromostyrene[23] (**12**) with phenylacetylene proceeded smoothly to give the corresponding 1,3-enyne product **13** in 52% yield. With our interest in the design of fluorescent compounds,[24] we attempted the synthesis of **14**, which possesses a strong π–π conjugated system, by the coupling reaction of 1,4-dibromobeneze (**15**) with phenylacetylene. However, compound **16** was obtained, as confirmed by NMR spectroscopic analysis and HRMS data. It is interesting to find that a decrease in the amount of phenylacetylene led to a higher yield of **16** (84%), whereas an increase in the amount of phenylacetylene (2.4 equiv.) only gave the product in 77% yield (Scheme 2).



12 1.0 equiv
1.2 equiv
13 52%

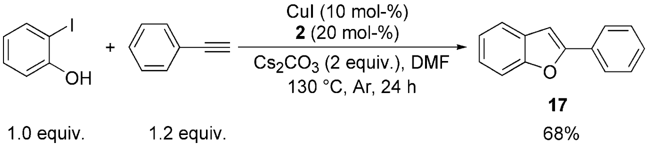
Scheme 1. Coupling between (*E*)-β-bromostyrene and phenylacetylene catalyzed by **2**/CuI.



15
1.0 equiv. 1.2 equiv. **16** 84%
1.0 equiv. 2.4 equiv. 77%

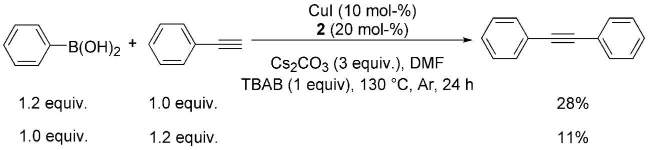
Scheme 2. Coupling between 1,4-dibromobenzene and phenylacetylene catalyzed by **2**/CuI.

Under the standard conditions, the coupling of 2-iodophenol with phenylacetylene was tested as shown in Scheme 3. To our delight, we did not obtain the Sonogashira coupling product but cyclic **17** instead in 68% yield. 2-Phenylbenzofuran derivatives such as **17** have been used as pharmaceuticals for the treatment and prophylaxis of bacterial infectious diseases or mycoses.[25]



Scheme 3. Coupling between 2-iodophenol and phenylacetylene catalyzed by **2**/CuI.

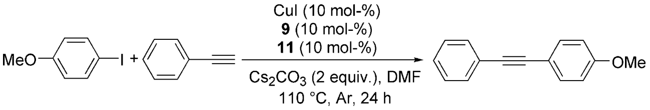
The typical Sonogashira coupling reaction is described as the reaction between aryl halides with alkynes. Here, we employed phenylboronic acids to replace the aryl halides. The desired coupling products between the phenylboronic acids and phenylacetylene were obtained in promising yield (Scheme 4).



Scheme 4. Coupling between phenylboronic acid and phenylacetylene catalyzed by **2**/CuI.

The CuI/8-hydroxyquinoline system represents a new bifunctional catalyst, which has been successfully used in alkynylation coupling. In this way, another possible bifunctional copper-catalyst has been designed and applied in this coupling reaction. As shown in Table 3, ligand **9** (10 mol-%) together with ligand **11** (10 mol-%) afforded the corresponding product from 4-iodoanisole with phenylacetylene in 74% yield (Table 3, Entry 1). An increase in the loading of the ligands led to good results (86%; Table 3, Entry 2). From these data it can be seen that the novel bifunctional catalyst showed obviously better catalytic efficiency than the ligand combined with CuI (Figure 2). This suggested the design of the bifunctional copper catalytic system for this coupling reaction, which is shown in Figure 3.

Table 3. The coupling of 4-iodoanisole and phenylacetylene by using another bifunctional copper-based catalytic system.[a]



Entry	Loading of **9** [mol-%]	Loading of **11** [mol-%]	*T* [°C]	*t* [h]	Yield [%][b]
1	10	10	130	24	74
2	20	20	130	24	86

[a] Reaction conditions: under an argon atmosphere, 4-iodoanisole (0.5 mmol), phenylacetylene (0.6 mmol), CuI (0.05 mmol), Cs_2CO_3 (1.0 mmol), DMF (2 mL), 130 °C. [b] Isolated yield (based on aryl halide).

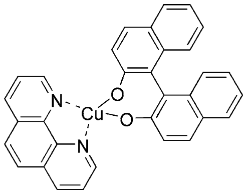


Figure 3. The novel bifunctional copper catalyst system was designed.

Conclusions

An inexpensive, mild, and efficient bifunctional CuI/**2** system for the alkynylation coupling reaction has been developed. In comparison to previous reports, the catalytic reaction is easy and generally applicable to a variety of aryl halides, heteroaryl halides, and vinyl halides, and this also provides an alternative to the palladium catalyst systems. Thus, the easy availability of the catalyst makes this catalytic method potentially useful, which could be amenable to scale up. Further work is in progress in this laboratory with the aim of extending the application of these readily available catalytic systems in other coupling transformations.

Experimental Section

General Experimental: All reactions were carried out under an argon atmosphere. Solvents were dried and degassed by standard methods, and all aryl and heteroaryl halides were purchased from Aldrich and Alfa. Phenylboronic acid and phenylacetylene are readily available. Flash column chromatography was performed by using silica gel (300–400 mesh). Analytical thin-layer chromatography was performed by using glass plates precoated with silica gel (200–400 mesh) impregnated with a fluorescent indicator (254 nm). NMR spectra were measured as $CDCl_3$ solutions with a Varian Inova-400 NMR spectrometer (400 MHz or 300 MHz) with TMS as an internal reference. Products were characterized by comparison of NMR spectroscopic and HRMS data with those in the literature.

Typical Experimental Procedure for the CuI/2 Catalyzed Alkynylation Coupling Reaction: A mixture of aryl halide (0.5 mmol), phenylacetylene (0.6 mmol), CuI (10 mol-%), **2** (20 mol-%), Cs_2CO_3 (2 equiv.), and DMF (2 mL) in a Schlenk tube was stirred under an argon atmosphere at 110–130 °C for the desired time until complete consumption of the starting material as monitored by TLC. The mixture was poured into ether, washed with water, extracted with ethyl acetate, dried with anhydrous $MgSO_4$, and then filtered. The solvent was evaporated under vacuum, and the residue was then purified by flash column chromatography (petroleum ether/ethyl acetate) to afford the corresponding coupled products.

Supporting Information (see footnote on the first page of this article): ^1H NMR spectra and HRMS data of the coupling products.

Acknowledgments

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