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Lewis Acid- and Organocatalyst-Cocatalyzed Multicomponent Reactions of 2-Alkynylbenzaldehydes, Amines, and Ketones

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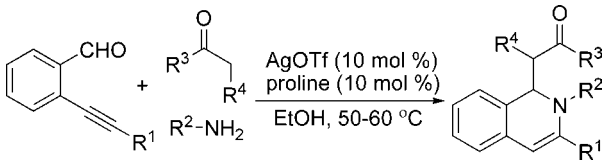
ABSTRACT



One-pot combination of silver triflate and proline catalysis is highly effective for the synthesis of 1,2-dihydroisoquinoline derivatives via multicomponent reactions of 2-alkynylbenzaldehydes, amines, and ketones.

The dual-activation concept (dual activation of the electrophiles and nucleophiles) is currently receiving considerable attention.[1] One of the general strategies of dual activation is that two separate catalysts can be combined in one catalytic system. In 2005, Yamamoto proposed "designer acids" to form a combination of acids with higher reactivity, selectivity, and versatility than the individual acid catalysts.[2] Recently, two-component activation systems that combine metal catalysis and the employment of stoichiometric or catalytic amounts of organocatalyst[3] have been successfully employed in allylic alkylation reactions.[4] For example, Córdova[4a] developed direct catalytic α-allylic alkylation of aldehydes and cyclic ketones via combination of palladium and enamine catalysis. Prompted by these results and our efforts for natural productlike compound synthesis,[5] we believed that the one-pot combination of metal and enamine catalysis may be applied for the synthesis of 1,2-dihydroisoquinoline derivatives via multicomponent reactions.[6] As outlined in Scheme 1, starting from 2-alkynylbenzaldehyde **1**, amine **2**, and ketone **3**, the merging of two powerful catalysts would enable both electrophilic and nucleophilic

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(1) For selected examples, see: (a) Ma, J.; Cahard, D. *Angew. Chem., Int. Ed.* **2004**, *43*, 4566. (b) Gröger, H. *Chem.-Eur. J.* **2001**, *7*, 5246. (c) Shibasaki, M.; Sasai, H.; Arai, T. *Angew. Chem., Int. Ed. Engl.* **1997**, *36*, 1236. (d) Matsui, K.; Takizawa, S.; Sasai, H. *Synlett* **2006**, 761. (e) Gryko, D.; Zimnicka, M.; Lipinski, R. *J. Org. Chem.* **2007**, *72*, 964.

(2) For a review, see: Yamamoto, H.; Futatsugi, K. *Angew. Chem., Int. Ed.* **2005**, *44*, 1924.

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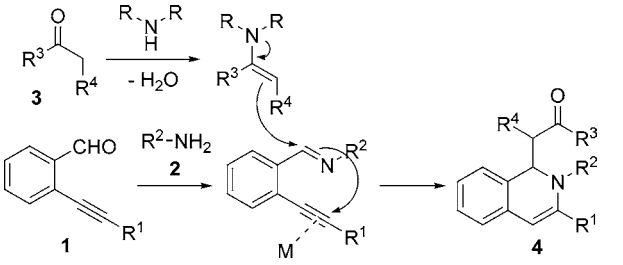
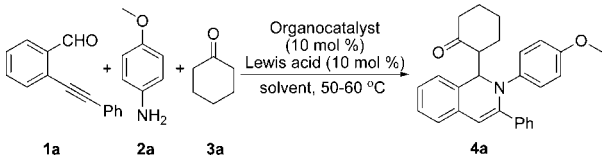
Scheme 1. Proposed 1,2-Dihydroisoquinoline Synthesis via Combination of Metal and Enamine Catalysis



activation, which is not fulfilled by one activation mechanism alone.

It is well-known that the 1,2-dihydroisoquinoline ring represents one of the most ubiquitous heterocyclic motifs and is found in many natural products and pharmaceuticals that exhibit remarkable biological activities.[7] Significant effort continues to be given to the development of new 1,2-dihydroisoquinoline-based structures and new methods for their construction,[8,9] due to their importance as substructures in a broad range of natural and designed products[7] as well as synthetic intermediates[10] in total synthesis of natural alkaloids. To verify the practicability of the projected route as shown in Scheme 1, we decided to pursue this highly challenging strategy: the one-pot combination of metal and enamine catalysis for the synthesis of 1,2-dihydroisoquinolines. Our studies commenced with the reaction of 2-alkynylbenzaldehyde **1a**,[10e,11] *p*-anisidine **2a**, and cyclohexanone **3a**, and the results are shown in Table 1.

Table 1. Conditions Screening for the Reaction of 2-Alkynylbenzaldehyde **1a**, *p*-Anisidine **2a**, and Cyclohexanone **3a** Catalyzed by Lewis Acid and Organocatalyst[a]



entry	Lewis acid (10 mol %)	organocatalyst (10 mol %)	solvent	time (h)	yield (%)[b]
1	$PdCl_2$	proline	EtOH	6	10
2	$PdCl_2(PhCN)_2$	proline	EtOH	6	11
3	$Pd(OAc)_2$	proline	EtOH	6	20
4	$Cu(OTf)_2$	proline	EtOH	6	15
5	$CuSO_4$	proline	EtOH	6	27
6	CuI	proline	EtOH	4	59
7	AgOTf	proline	EtOH	4	65
8	AgOTf	proline	MeOH	4	65
9	AgOTf	proline	toluene	24	22
10	AgOTf	proline	THF	24	25
11	AgOTf	proline	DCE	24	13
12	AgOTf	$^{i}Pr_2NH_2$	EtOH	4	52
13	AgOTf	pyrrolidine	EtOH	4	41
14	AgOTf	piperidine	EtOH	4	40
15	AgOTf	proline[c]	EtOH	4	65
16	AgOTf	–	EtOH	24	18
17	AgOTf[d]	proline	EtOH	8	42
18	AgOTf[e]	proline	EtOH	8	32
19[f]	AgOTf	proline	EtOH	4	40

[a] Reaction conditions: 2-alkynylbenzaldehyde **1a** (0.30 mmol), *p*-anisidine **2a** (1.0 equiv), cyclohexanone **3a** (5.0 equiv), Lewis acid (10 mol %), organocatalyst (10 mol %), solvent (2.0 mL), 50−60 °C. [b] Isolated yield based on 2-alkynylbenzaldehyde **1a**. [c] 30 mol % of proline was utilized.[d] 5 mol % of AgOTf. [e] 2.5 mol % of AgOTf. [f] 2.5 equiv of cyclohexanone **3a** was employed.

Soft metal salts such as palladium, copper, or silver with mild Lewis acidity were examined for the reaction of 2-alkynylbenzaldehyde **1a**, *p*-anisidine **2a**, and cyclohexanone **3a**. Gratifyingly, in an initial experiment, we observed the

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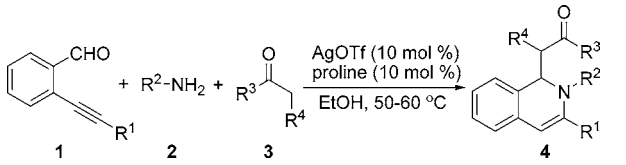
formation of the desired product **4a** (10% yield) when the reaction was performed in EtOH catalyzed by $PdCl_2$ (10 mol %) and proline (10 mol %) (Table 1, entry 1). Further screening of metal salts revealed that the yield could be dramatically improved when CuI or AgOTf combined with proline was utilized in the reaction (59% and 65% yield, respectively; Table 1, entries 6 and 7). A similar result was obtained when the solvent was changed to methanol (Table 1, entry 8). Inferior results were displayed when other solvents were used in the reaction. We also tried other secondary amines for enamine generation, and proline was demonstrated to be the best choice (Table 1, entries 12−14). Increasing the amount of proline catalyst could not improve the yield (Table 1, entry 15). Only 18% of the corresponding product **4a** was generated in the absence of proline catalyst (Table 1, entry 16). A reduced yield was observed when the amount of AgOTf was decreased (Table 1, entries 17 and 18). Five equivalents of cyclohexanone **3a** is essential in the reaction to obtain a respectable yield (Table 1, entry 19). In these reactions, a small amount of diastereoisomer was detected (<15% yield). Although chiral proline was utilized as catalyst, only the racemic product was generated via chiral HPLC determination (please see Supporting Information).

To demonstrate the generality of this method, the scope of the reaction was investigated under optimized conditions [AgOTf (10 mol %), proline (10 mol %), EtOH, 50−60 °C], and the results are summarized in Table 2. From Table 2, we found that the conditions have proven to be useful for a range of 2-alkynylbenzaldehydes, amines, and ketones, and these reactions usually went to completion in 2 h. For reaction of 2-alkynylbenzaldehyde **1a**, anilines, and cyclohexanone **3a**, both electron-rich and electron-poor anilines, which are suitable partners in this process, give similar yields (Table 2, entries 1−3). When cyclohexanone **3a** was replaced by butan-2-one (**3b**) in the reaction of 2-alkynylbenzaldehyde **1a** with anilines, it was found that a range of substitutions in the anilines were well tolerated under our conditions (Table 2, entries 4−10). Thus, *p*-anisidine **2a** or *p*-toluidine **2b** reacted with 2-alkynylbenzaldehyde **1a** and butan-2-one (**3b**) to give the desired 1,2-dihydroisoquinoline **4d** or **4e** in 80% or 91% yield, respectively (Table 2, entries 4 and 5). Similarly, the reaction of 2-alkynylbenzaldehyde **1a**, aniline **2c**, and butan-2-one **3b** furnished the expected product **4f** in 82% yield (Table 2, entry 6). 2-Alkynylbenzaldehyde **1a** reacted smoothly with butan-2-one (**3b**) and 4-chloroaniline (**2d**) or 4-fluoroaniline (**2e**) leading to the corresponding product **4g** or **4h** in 95% or 77% yield, respectively (Table 2, entries 7 and 8). 3-Nitroaniline (**2f**) or 3-(trifluoromethyl)-aniline (**2g**) was also employed in the reaction, affording

Table 2. Reaction of 2-Alkynylbenzaldehyde **1**, Amine **2**, and Ketone **3** Catalyzed by Silver Triflate and Proline[12]



entry	R^1	R^2	R^3/R^4	yield (%)[a]
1	C_6H_5 (**1a**)	4-$MeOC_6H_4$ (**2a**)	$-(CH_2)_4-$ (**3a**)	65 (**4a**)[b]
2	C_6H_5 (**1a**)	C_6H_5 (**2c**)	$-(CH_2)_4-$ (**3a**)	61 (**4b**)[c]
3	C_6H_5 (**1a**)	4-FC_6H_4 (**2e**)	$-(CH_2)_4-$ (**3a**)	60 (**4c**)[d]
4	C_6H_5 (**1a**)	4-$MeOC_6H_4$ (**2a**)	C_2H_5/H (**3b**)	80 (**4d**)
5	C_6H_5 (**1a**)	4-MeC_6H_4 (**2b**)	C_2H_5/H (**3b**)	91 (**4e**)
6	C_6H_5 (**1a**)	C_6H_5 (**2c**)	C_2H_5/H (**3b**)	82 (**4f**)
7	C_6H_5 (**1a**)	4-ClC_6H_4 (**2d**)	C_2H_5/H (**3b**)	95 (**4g**)
8	C_6H_5 (**1a**)	4-FC_6H_4 (**2e**)	C_2H_5/H (**3b**)	77 (**4h**)
9	C_6H_5 (**1a**)	3-$NO_2C_6H_4$ (**2f**)	C_2H_5/H (**3b**)	78 (**4i**)
10	C_6H_5 (**1a**)	3-$CF_3C_6H_4$ (**2g**)	C_2H_5/H (**3b**)	68 (**4j**)
11	C_6H_5 (**1a**)	$C_6H_5CH_2$ (**2h**)	C_2H_5/H (**3b**)	32 (**4k**)
12	C_6H_5 (**1a**)	4-$MeOC_6H_4$ (**2a**)	CH_3/H (**3c**)	77 (**4l**)
13	C_6H_5 (**1a**)	C_6H_5 (**2c**)	CH_3/H (**3c**)	77 (**4m**)
14	C_6H_5 (**1a**)	4-FC_6H_4 (**2e**)	CH_3/H (**3c**)	93 (**4n**)
15	C_6H_5 (**1a**)	4-$MeOC_6H_4$ (**2a**)	$(CH_2)_2CH_3$/H (**3d**)	80 (**4o**)
16	C_6H_5 (**1a**)	4-$MeOC_6H_4$ (**2a**)	4-$MeOC_6H_4$/H (**3e**)	61 (**4p**)
17	C_6H_5 (**1a**)	4-$MeOC_6H_4$ (**2a**)	CH_2COCH_3/H (**3f**)	60 (**4q**)
18	C_6H_5 (**1a**)	4-$MeOC_6H_4$ (**2a**)	CH_2CH_3/CH_3 (**3g**)	30 (**4r**)[e]
19	4-$MeOC_6H_4$ (**1b**)	4-$MeOC_6H_4$ (**2a**)	C_2H_5/H (**3b**)	91 (**4s**)
20	4-$MeOC_6H_4$ (**1b**)	4-FC_6H_4 (**2e**)	C_2H_5/H (**3b**)	50 (**4t**)
21	cyclopropyl (**1c**)	4-$MeOC_6H_4$ (**2a**)	C_2H_5/H (**3b**)	30 (**4u**)
22	cyclopropyl (**1c**)	4-FC_6H_4 (**2e**)	C_2H_5/H (**3b**)	35 (**4v**)
23	*n*-Bu (**1d**)	4-$MeOC_6H_4$ (**2a**)	C_2H_5/H (**3b**)	28 (**4w**)
24	$SiMe_3$ (**1e**)	4-$MeOC_6H_4$ (**2a**)	C_2H_5/H (**3b**)	12 (**4x**)[f]

[a] Isolated yield based on 2-alkynylbenzaldehyde **1**. [b] Along with 15% of diastereoisomer **4a′**. [c] Along with 25% of diastereoisomer **4b′**. [d] Along with 27% of diastereoisomer **4c′**. [e] Along with 20% of diastereoisomer **4r′**. [f] R^1 = H in compound **4x**.

the expected product in good yield (Table 2, entries 9 and 10). However, a low yield (32%) was observed when an aliphatic amine such as benzylamine was utilized in the reaction of 2-alkynylbenzaldehyde **1a** and butan-2-one (**3b**) (Table 2, entry 11). Interestingly, the above reactions were highly regiospecific for this nonsymmetric ketone **3b**, and only one isomer was generated in the reaction. This phenomenon was also observed for substrates **3d**, **3e**, and **3f**. Acetone (**3c**) was also a good partner in this multicomponent reaction (Table 2, entries 12−14). 3-Pentanone (**3g**) was then examined for the reaction of 2-alkynylbenzaldehyde **1a** and *p*-anisidine **2a**. As expected, a mixture of diasteromers was generated (Table 2, entry 18). When 2-alkynylbenzaldehyde **1b** was used as a substrate for the reaction with butan-2-one (**3b**), anilines with electron-donating groups gave much better results. For example, *p*-anisidine **2a** reacted with 2-alkynylbenzaldehyde **1b** and butan-2-one (**3b**) to give the desired 1,2-dihydroisoquinoline **4s** in 91% yield (Table 2, entry 19), whereas a 50% yield of product **4t** was afforded when 4-fluoroaniline (**2e**) was utilized in the same reaction (Table 2, entry 20). However, low yields were obtained when other 2-alkynylbenzaldehydes (such as **1c** and **1d**) reacted with anilines and butan-2-one (**3b**). In the case of substrate **1e** (R^1 = $SiMe_3$), only desilyl product **4x** was generated (Table 2, entry 24). As described above, the optimal active

(12) General procedure for one-pot reaction of 2-alkynylbenzaldehyde **1**, amine **2**, and ketone **3** catalyzed by silver triflate and proline: A solution of 2-alkynylbenzaldehyde **1** (0.30 mmol), amine **2** (0.30 mmol, 1.0 equiv), ketone **3** (1.50 mmol, 5.0 equiv), AgOTf (0.03 mmol, 10 mol %), proline (0.03 mmol, 10 mol %) in C_2H_5OH (1.5 mL) was stirred at 50−60 °C under N_2 for a period of time. After completion of the reaction as indicated by TLC, the solvent was evaporated and then quenched with water (10 mL), extracted with EtOAc (2 × 10 mL), and dried by anhydrate Na_2SO_4. Evaporation of the solvent followed by purification on silica gel provided the corresponding product **4**. (For details, please see Supporting Information.)

products might be generated during the reaction process in these reactions since a chiral catalyst (proline) was used in the reaction. Compound **4l** was selected for chiral HPLC analysis, again, no enantioselectivity was observed.

In summary, we have described a novel and highly effective one-pot combination of AgOTf and proline catalysis for the synthesis of 1,2-dihydroisoquinoline derivatives via multicomponent reactions of 2-alkynylbenzaldehydes, amines, and ketones. These results not only represent an efficient example of Lewis acid−organocatalyst combination in organic synthesis, but also provide a facile and efficient route to 1,2-dihydroisoquinoline derivatives, which could be directly used for biological assays. Application of metal catalysis−organocatalyst combination for other transformations is currently under investigation in our research group, which will be reported in due course.

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Supporting Information Available: Experimental procedures, characterization data, copies of ^{1}H and ^{13}C NMR of new compounds. This material is available free of charge via the Internet at http://pubs.acs.org.

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