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Facile, Mild, and Highly Enantioselective Alkynylzinc Addition to Aromatic Aldehydes by BINOL/*N*-Methylimidazole Dual Catalysis

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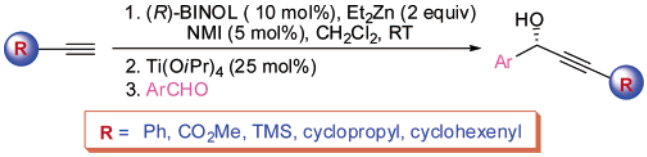

The dual Lewis acid/base catalytic system, generated from *N*-methylimidazole (NMI), (*R*)-1,1′-bi-2-naphthol [(*R*)-BINOL], and Ti(O*i*Pr)$_4$, effectively catalyzes the enantioselective alkynylation of aldehydes in the presence of Et$_2$Zn in good yields and excellent enantioselectivities of up to 94% ee at room temperature. The mild reaction conditions make it possible to use functional alkynes in this asymmetric addition.

The catalytic asymmetric alkynylzinc addition to aldehydes can provide a very convenient route to the corresponding chiral secondary propargylic alcohols,[1] which have been identified as versatile building blocks for fine chemicals, pharmaceuticals, and natural products.[2] It is therefore not surprising that great efforts have recently been directed toward the development of this important asymmetric reaction, and two general catalytic systems are currently considered to be the most practical. One has recently been discovered by Carreira and co-workers, using stoichiometric or catalytic quantities of Zn(OTf)$_2$, *N*-methylephedrine, and Et$_3$N to afford the desired products in high yields and enantioselectivities in the addition of terminal acetylenes to aldehydes.[3] In this approach, the zinc alkynylides are generated in situ from terminal alkynes and Zn(OTf)$_2$ in the presence of amine base.[3,4] Another system involves the use of Et$_2$Zn or Me$_2$Zn to prepare in situ the alkynylzincs.[5−8] In this respect, direct use of the readily available and inexpensive unmodified 1,1′-bi-2-naphthol (BINOL) in combination with Ti(O*i*Pr)$_4$ and Et$_2$Zn or Me$_2$Zn has resulted in a series of exciting findings.[9,10]

BINOL−Ti complex can catalyze alkyne addition to alkyl, aryl, and α,β-unsaturated aldehydes with high ee values and good yields in the presence of Et$_2$Zn.[9a,b] However, in these systems, higher reaction temperatures are generally required in the first step to prepare the corresponding alkynylzinc from the terminal alkyne and alkylzinc reagent.[9a−c] The high temperature for the preparation of the alkynylzinc reagents may cause the decomposition of certain functional alkynes. Although the use of Me$_2$Zn instead of Et$_2$Zn can lower the reaction temperature in the formation of alkynylzinc, these BINOL−Ti(O*i*Pr)$_4$−Me$_2$Zn systems generally require a separate step to synthesize alkynylzinc.[9d,10]

Quite recently, Pu et al. discovered that the addition of hexamethylphosphoramide (HMPA) greatly accelerates the reaction of Et$_2$Zn with terminal alkynes at room temperature while maintaining the high enantioselectivity for the addition to aldehydes.[11] The mild condition for the formation of alkynylzinc reagents avoids the reflux of the toluene solutions of the alkynes and Et$_2$Zn as previously reported[9a−c] and enables the use of functional alkynes in this asymmetric reaction with high enantioselectivity. However, the protocol requires substoichiometric quantities of chiral BINOL (40 mol %), 2 equiv

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(4) The use of stoichiometric amounts of chiral amino alcohol-based ligands: (a) Jiang, B.; Chen, Z. L.; Xiong, W. N. *Chem. Commun.* **2002**, 1524. (b) Chen, Z. L.; Xiong, W. N.; Jiang, B. *Chem. Commun.* **2002**, 2098. (c) Jiang, B.; Si, Y. G. *Adv. Synth. Catal.* **2004**, *346*, 669.

(5) Bisoxazolidine-catalyzed enantioselective alkynylation of aromatic aldehydes at low temperature (−4 to −15 °C): Wolf, C.; Liu, S. *J. Am. Chem. Soc.* **2006**, *128*, 10996.

(6) Catalytic procedures based on amino alcohol ligands focus on the addition of phenylacetylene to aromatic aldehydes, while few address the need to utilize functional alkynes: (a) Xu, Z.; Wang, R.; Xu, J.; Da, C.; Yan, W.; Chen, C. *Angew. Chem., Int. Ed.* **2003**, *42*, 5747. (b) Xu, Z.; Chen, C.; Xu, J.; Miao, M.; Yan, W.; Wang, R. *Org. Lett.* **2004**, *6*, 1193. (c) Dahmen, S. *Org. Lett.* **2004**, *6*, 2113. (d) Fang, T.; Du, D. M.; Lu, S. F.; Xu, J. *Org. Lett.* **2005**, *7*, 2081.

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(8) Selected examples for modified BINOLs as ligands: (a) Moore, D.; Huang, W. S.; Xu, M. H.; Pu, L. *Tetrahedron Lett.* **2002**, *43*, 8831. (b) Xu, M. H.; Pu, L. *Org. Lett.* **2002**, *4*, 4555. (c) Liu, Q. Z.; Xie, N. S.; Luo, Z. B.; Cui, X.; Cun, L. F.; Gong, L. Z.; Mi, A. Q.; Jiang, Y. Z. *J. Org. Chem.* **2003**, *68*, 7921. (d) Liu, L.; Pu, L. *Tetrahedron* **2004**, *60*, 7427. (e) Li, Z. B.; Pu, L. *Org. Lett.* **2004**, *6*, 1065.

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(10) The method also required 1 equiv of an additional chiral sulfonamide ligand and was only applied in the addition of phenylacetylene to aromatic aldehydes below 0 °C for 1−2 days: Li, X. S.; Lu, G.; Kwok, W. H.; Chan, A. S. C. *J. Am. Chem. Soc.* **2002**, *124*, 12636.

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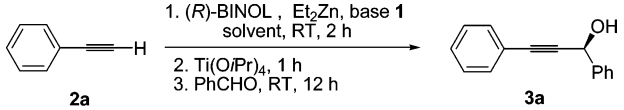
of HMPA, 1 equiv of Ti(O*i*Pr)$_4$, and 4 equiv of Et_2Zn to achieve excellent enantioselectivities. Clearly, it is highly desirable to reduce loadings of chiral BINOL as well as other additives to make this process practical and economical. This should also represent a partial solution to a long-standing challenge in asymmetric catalysis.[12]

Recently, the concept of dual acid/base catalysis has been introduced for asymmetric catalytic reactions, in which both electrophilic and nucleophilic precursors are activated by catalytic quantities of chiral Lewis acid and base, respectively.[13] Imidazole (Im) and its derivatives are ubiquitous in biological and biochemical structure and function and play the key roles in the synergistically catalytic mechanisms of many enzymes. Recently, some examples have demonstrated that imidazoles can be employed as the base moiety of dual activation or co-catalysis for the asymmetric catalytic reaction,[14] and the basic nitrogen of the imidazole ring can activate the alkyne.[15] Thus we speculated that application of a double catalytic activation strategy seems to be one of the most promising solutions, which should cause the chiral BINOL−Ti complex and Im to activate electrophiles (carbonyl compounds) and nucleophiles (terminal alkynes), respectively. Following our continuing interest in the development of dual catalysis[16] as well as imidazole chemistry,[17] we therefore focused on dual activation based on imidazole as well as its derivatives. It is reasonable to assume that imidazoles **1** might meet the requirement as Lewis bases to facilitate the deprotonation of terminal acetylenes to improve the formation of alkynylzinc reagents,[18] as we had observed that benzaldehyde was able to react smoothly with phenylacetylene with the assistance of 5 mol % of *N*-methylimidazole (NMI) **1a** and 2.0 equiv of Et_2Zn at room temperature with the formation of the racemic alkynylation product in 52% yield (Table 1, entry 1). As expected, we herein report that the truly BINOL−Ti catalytic version of alkyne addition to aldehyde could be achieved with high enantioselectivity in the presence of Et_2Zn at room temperature when a catalytic amount of NMI was employed as Lewis base.[11]

Initial studies on the development of the asymmetric reaction conditions revealed that the quantity of NMI was crucial to the outcome of the reaction, and the effect of the chiral Lewis acid/base ratio is described in Table 1, which shows a clear maximum at 5 mol % of NMI in combination with 10 mol % of (*R*)-BINOL, 25 mol % of Ti(O*i*Pr)$_4$, and 2 equiv of Et_2Zn in CH_2Cl_2 at room temperature (Table 1, entry 3). Further increasing the quantity of NMI might lead to diminished enantioselectivity, which should be attribute to the competition between the dual catalysis path and the Lewis base (only NMI) catalysis path, although the chemical yield of propargylic alcohol could be improved slightly (compare entries 1−3 in Table 1). These results show that our optimized catalytic system significantly overcomes troubles potentially arising from the chemical

TABLE 1. Some Representative Results from the Screening of Reaction Conditions for the Additions of Phenylacetylene with Benzaldehyde in the Presence of (*R*)-BINOL, Ti(O*i*Pr)$_4$, Et_2Zn, and Im[a]

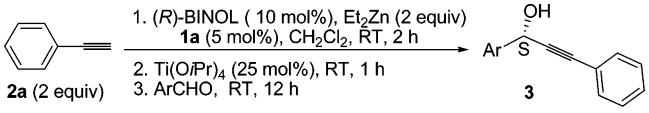


2a **3a**

entry	base (mol %)[b]	BINOL (mol %)	BINOL/Ti(O*i*Pr)$_4$	solvent	yield (%)[c]	ee (%)[d]
1	**1a** (5)			CH_2Cl_2	52	
2	**1a** (10)	10	1:2.5	CH_2Cl_2	94	82
3	**1a** (5)	10	1:2.5	CH_2Cl_2	92	93
4	**1a** (2.5)	10	1:2.5	CH_2Cl_2	72	91
5[e]	**1a** (5)	10	1:2.5	CH_2Cl_2	83	90
6	**1a** (5)	10	1:2	CH_2Cl_2	54	83
7	**1a** (5)	10	1:3	CH_2Cl_2	92	89
8	**1a** (5)	10	1:5	CH_2Cl_2	93	82
9	**1a** (5)	10	1:2.5	THF	68	79
10	**1a** (5)	10	1:2.5	Et_2O	ND	60
11	**1a** (5)	10	1:2.5	toluene	ND	76
12	**1a** (2.5)	5	1:2.5	CH_2Cl_2	68	84
13	**1b** (5)	10	1:2.5	CH_2Cl_2	83	87
14	**1c** (5)	10	1:2.5	CH_2Cl_2	78	87
15	**1d** (5)	10	1:2.5	CH_2Cl_2	71	84

[a] Et_2Zn/phenylacetylene/aldehyde = 2:2:1. [b] NMI (**1a**), imidazole (**1b**), bisglyoxaline (**1c**), benzimidazole (**1d**). [c] Yield of isolated product based on aldehyde after chromatographic purification. [d] Enantiomeric excess was determined by chiral HPLC analysis (Chiralcel OD-H). [e] The formation time of zinc phenylacetylide = 1 h.

TABLE 2. Scope of Asymmetric Phenylacetylene Addition to Aromatic Aldehydes



2a (2 equiv) **3**

entry	aldehyde (Ar)	product	yield (%)[a]	ee (%)[b]
1	Ph	**3a**	92	93
2	2-MeC_6H_4	**3b**	91	93
3	3-MeC_6H_4	**3c**	92	94
4	4-MeC_6H_4	**3d**	81	92
5	3-ClC_6H_4	**3e**	95	92
6	4-ClC_6H_4	**3f**	80	92
7	4-BrC_6H_4	**3g**	85	92
8	4-FC_6H_4	**3h**	91	93
9	4-MeOC_6H_4	**3i**	87	93
10	3-MeOC_6H_4	**3j**	82	91
11	2-furyl	**3k**	92	87
12	2-naphthyl	**3l**	85	93

[a] Yield of isolated product based on aldehyde after chromatographic purification. [b] Enantiomeric excess was determined by chiral HPLC analysis (Chiralcel OD-H). The absolute configurations of adducts were assigned by comparison with literature data.

(12) In this regard, one impressive example has been illustrated by the recent studies of the Carreira group, in which a catalytic version of the addition procedure could be conducted at elevated temperatures, i.e., 60 °C, to overcome the kinetic barrier inhibiting protonation of the first formed Zn-alkoxide, avoiding the use of stoichiometric quantities of $Zn(OTf)_2$ and *N*-methylephedrine. See ref 3b.

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(16) Ma, K. Y.; You, J. S. *Chem.—Eur. J.* **2007**, *13*, 1863−1871.

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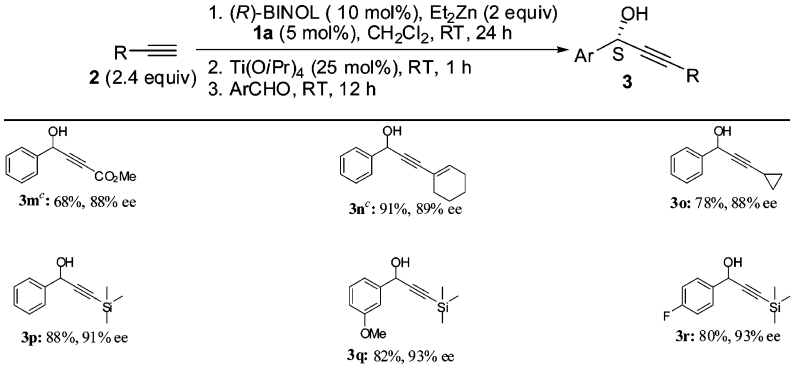
TABLE 3. Catalytic, Enantioselective Addition of Functional Alkynes to Aromatic Aldehydes[a,b]



[a] The time of alkynylzinc formation = 24 h. [b] Yield of isolated product based on aldehyde after chromatographic purification. Enantiomeric excess was determined by chiral HPLC analysis (Chiralcel OD-H or AD-H). The absolute configurations of adducts were assigned by comparison with literature data. [c] 2.1 equiv of RCCH.

incompatibility of Lewis acids and bases and suppresses the NMI-initiated background reaction. Furthermore, a series of imidazoles (e.g., imidazole **1b**, bisglyoxaline **1c**, and benzimidazole **1d**) were also tested in the reaction between benzaldehyde and phenylacetylene, indicating NMI gave the best result (Table 1, entries 3, 13−15).

We found that this reaction was strongly influenced by the solvent: superior results could be obtained with CH_2Cl_2, while low enantioselectivities were found in toluene, diethyl ether, and THF (Table 1, entries 3, 9−11). We then varied the amount and ratio of both (*R*)-BINOL and Ti(O*i*Pr)$_4$ and found that the best ee value was obtained when the (*R*)-BINOL/Ti(O*i*Pr)$_4$ ratio is 1/2.5 (Table 1, entries 3, 6−8). It is noteworthy that 10 mol % of (*R*)-BINOL proved to be adequate in terms of both reactivity and selectivity (compare entries 3 and 12 in Table 1). In addition, no significant changes in ee values were observed when the reaction temperature was decreased from room temperature to 0 °C, but it slowed down the reaction rate and reduced the yield of the product.

Having optimized the (*R*)-BINOL/NMI-catalyzed alkynylation of benzaldehyde with phenylacetylene, we decided to screen a series of aromatic aldehydes and acetylenes to evaluate the scope of this reaction. We were delighted to find that alkynylation of electron-rich and -deficient aromatic aldehydes with phenylacetylene gave the corresponding propargylic alcohols in excellent ee values (87−94% ee) and yields (80−95%) at room temperature (Table 2). It is noteworthy that the enantioselectivity of the reaction seems to be independent of the substitution of the aromatic aldehyde. Thus this catalytic process displays relatively wide substrate scope, and the presented results are comparable to those attained by substoichiometric and/or stoichiometric amounts of BINOL−Ti(O*i*Pr)$_4$−HMPA−Et_2Zn as previously reported by Pu and co-workers.[11a]

Furthermore, the optimized conditions were also applicable to some quite challenging alkynes for existing catalytic systems, as summarized in Table 3. For example, ethynylcyclohexene and cyclopropylacetylene instead of phenylacetylene, both good chemical yields and excellent enantioselectivities, were achieved, although a longer time of alkynylzinc formation (24 h) was required, which compared to the results obtained with Wolf's bisoxazolidine catalyst system (Table 3, **3n**−**3o**).[5] Trimethylsilylacetylene is a particularly desirable alkyne due to the possible use of the desilylated product for alkylation and the Sonogashira coupling.[19] To our delight, our catalytic system could also be employed in the asymmetric addition of trimethylsilylacetylene to aldehydes. The corresponding 3-silylpropargylic alcohols were obtained in up to 88% yield and 93% ee (Table 3, **3p**−**3r**).[20] γ-Hydroxy-α,β-acetylenic esters are very useful in the synthesis of highly functionalized organic molecules.[21] Noteworthy, our dual catalytic system could also be used for this asymmetric addition of methyl propiolate with benzaldehyde with high enantioselectivity (Table 3, **3m**).

In summary, we report the facile and mild highly enantioselective alkynylation of aldehydes using the dual acid/base catalysis, in which the BINOL/Ti(O*i*Pr)$_4$ complex would activate aldehydes while NMI would promote the deprotonation of terminal alkynes at room temperature. Therefore, the addition of catalytic quantities of NMI greatly reduced the temperature for the formation of the alkynylzinc reagents and significantly improved the previous BINOL/Et_2Zn/Ti(O*i*Pr)$_4$ system.[11] Thus the mild reaction conditions make it possible to use some quite challenging alkynes, such as ethynylcyclohexene, cyclopropylacetylene, trimethylsilylacetylene, and methyl propiolate, in this asymmetric addition. To the best of our knowledge, our dual acid/base catalytic system is among the most effective systems so far reported for the asymmetric alkynylzinc addition to aromatic aldehydes. The easy availability of the chiral ligand and NMI as well as the metal reagents and the mild reaction conditions make this process very practical.

Experimental Section

General Procedure for the Catalytic Enantioselective Addition of Alkyne to Aldehyde. Under nitrogen, to a flame-dried test

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(20) There was not any information describing the asymmetric addition of trimethylsilylacetylene to aldehydes in the BINOL−Ti(O*i*Pr)$_4$−HMPA−Et_2Zn as previously reported by Pu and co-workers. See ref 11.

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tube containing (*R*)-BINOL (7.2 mg, 0.025 mmol) and 3 mL of CH_2Cl_2 were sequentially added NMI (0.0125 mmol, 1 μL), alkyne (0.5 mmol), and Et_2Zn (0.5 mmol, 1.25 M in toluene). The resulting mixture was stirred at room temperature (20−23 °C) for 2 h, followed by the addition of $Ti(OiPr)_4$ (0.0625 mmol, 18.5 μL). After 1 h of stirring, an aldehyde (0.25 mmol) was added, and the resulting mixture was stirred for an additional 12 h. Saturated NH_4Cl solution (5 mL) was added to quench the reaction, and the mixture was extracted with ethyl acetate. The combined organic layers were dried over anhydrous $MgSO_4$, and solvents were removed under reduced pressure. The residue was purified by flash column chromatography (silica gel, 10% EtOAc in petroleum ether) to give the product.

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Supporting Information Available: Experimental procedures and full spectroscopic data for all compounds. This material is available free of charge via the Internet at http://pubs.acs.org.

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