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Direct C–N Coupling of Imidazoles with Aromatic and Benzylic Compounds via Electrooxidative C–H Functionalization

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Supporting Information

ABSTRACT: A method for the C–N coupling of imidazoles based on electrooxidative C–H functionalization of aromatic and benzylic compounds has been developed. The key to the success is the formation of protected imidazolium ions as initial products, avoiding overoxidation. Deprotection under nonoxidative conditions affords N-substituted imidazoles. Various functional groups are compatible with the present transformation. To demonstrate the power of the method, a P450 17 inhibiter and an antifungal agent having N-substituted imidazole structures were synthesized.

Imidazoles bearing organic substituents on the nitrogen atom are important motifs in natural products and medicinal compounds of various biological activities such as antiviral, anti-inflammatory, anticancer, and antifungal activities (Figure 1).

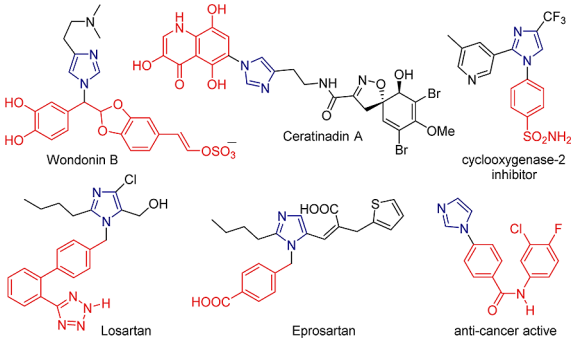

Figure 1. Selected examples of N-substituted imidazoles.

Some N-substituted imidazoles are widely used as drugs.[1] For example, losartan is the first orally available angiotensin II receptor antagonist and is used for treating hypertension.[2]

Although various protocols for C–N coupling of imidazole derivatives and organic molecules have been developed, most of them require prefunctionalization of organic molecules.[3−5] Because direct C–N coupling[6] between imidazoles and organic compounds serves as a straightforward method for construction of a wide variety of N-substituted imidazoles having intriguing chemical and biological functions, development of efficient methods for direct C–N coupling by C–H functionalization is highly desired. Recently, direct C–N coupling by functionalizing benzylic C–H or a C–H bond adjacent to a heteroatom based on radical H-atom abstraction has been reported.[7] Although it serves as a powerful method for synthesizing *N*-benzyl imidazoles, an excess amount of a benzylic compound is required presumably because C–N coupling products are also susceptible to H-atom abstraction.

Electrochemical oxidation[8,9] serves as a powerful method for functionalizing C–H bonds of benzylic[10] and aromatic[11] compounds by single electron transfer. Despite the usefulness of the method, it suffers from overoxidation when a product is also susceptible to electrochemical oxidation.[12] This is indeed the case when benzylic and aromatic compounds are electrochemically oxidized in the presence of unprotected imidazole (Figure 2a).

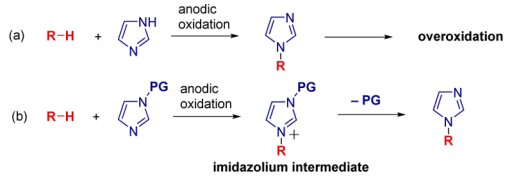

Figure 2. Electrooxidative C–N cross-coupling of imidazoles with benzylic and aromatic compounds. (a) Conventional approach. (b) Present approach.

Here, we report a new approach to solve the problem; the C–N coupling of protected imidazoles based on electrooxidative C–H functionalization of organic counterparts (Figure 2b). This method is applicable not only to benzylic but also to aromatic compounds. The initial products are *N*-benzyl or *N*-arylimidazolium ions, which can be easily converted to the corresponding *N*-benzyl or *N*-aryl imidazoles by a subsequent nonoxidative removal of the protecting group. The key to the success of the present approach is the intermediacy of the electrooxidatively inactive imidazolium ions, which avoids overoxidation. Therefore, the reaction does not require an excess amount of a benzylic or aromatic compound.

We first chose to use naphthalene (**1a**) because C–H functionalization of **1a** via the electrogenerated radical cation intermediate is well known.[13] Because the choice of an appropriate protecting group of an imidazole is crucial for the success of the present approach, the electrochemical reactions of naphthalene in the presence of various N-protected imidazoles were examined. The electrochemical reactions

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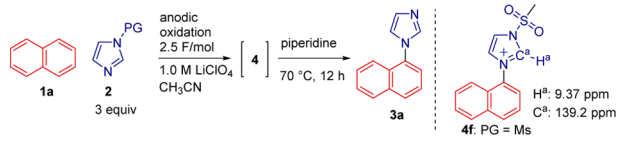
Table 1. Optimization of the Protecting Groups



entry	protecting group	protecting group label	yield of **3a** (%)[a]	entry	protecting group	protecting group label	yield of **3a** (%)[a]
1	H	**2a**	0	5	$SO_2C_6H_4$-*p*-CH_3 (Ts)	**2e**	71
2	$COCH_3$	**2b**	46				
3	CO_2NMe_2	**2c**	62	6	SO_2CH_3 (Ms)	**2f**	82
4	SO_2NMe_2	**2d**	42	7	$SiMe_3$	**2g**	0

[a]Yields were determined by GC analysis using tridecane as an internal standard.

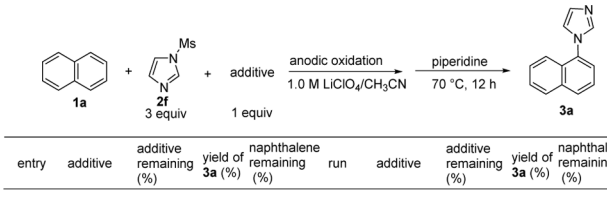
were carried out with 0.20 mmol of naphthalene (**1a**) and 0.6 mmol of a N-protected imidazole (**2**) in an H-type divided cell equipped with an anode consisting of fine carbon fibers and a platinum plate cathode using a 1.0 M solution of $LiClO_4$ in CH_3CN at 20 °C under constant current conditions. In the anodic chamber, **1a** was oxidized, and in the cathodic chamber, the proton was reduced to generate hydrogen. Trifluoromethanesulfonic acid was added to the cathodic chamber prior to the electrolysis in order to promote the hydrogen generation. After 2.5 F/mol of electricity was consumed, the reaction mixture was treated with piperidine at 70 °C for 12 h. The yields of the resulting C−N coupling product **3a** were determined by GC analysis.

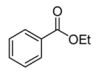
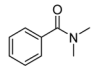
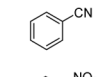
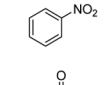
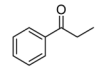
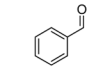
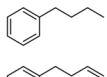
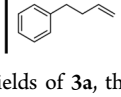
The use of unprotected imidazole **2a** gave no C−N coupling product presumably because of the overoxidation (Table 1, entry 1). In contrast, 1-acetylimidazole (**2b**) gave the C−N coupling product **3a** in 46% yield (entry 2). Among the examined groups (entries 2−7), *N*-methylsulfonyl-protected imidazole **2f** (1-mesylimidazole) gave the product in the highest yield (entry 6, 82%). The imidazolium intermediate **4f** could be characterized by NMR after electrolysis (see the Supporting Information).

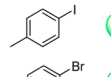
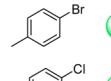
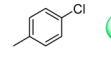
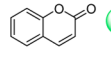
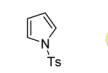
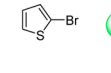
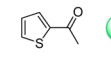
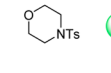
Various functional groups are compatible with the present transformation. To demonstrate high chemoselectivity of the present method, we applied a robustness screen that was recently developed by Glorius group.[14] The reactions were performed in the presence of various compounds, having given functional groups as additives. The conversion of naphthalene (**1a**), the yield of the product **3a**, and those of the unchanged additives were determined by GC analysis (Table 2). Electron-withdrawing groups such as ketone, ester, amide, cyano, and nitro groups survived the reaction (entries 2−4), although aldehydes could not tolerate the reaction conditions (entry 5). Benzylic C−H bonds in non-electron-rich aromatic rings and allylic C−H bonds were intact (entry 6). The observed chemoselectivity sharply contrasts with that for the radical H-atom abstraction.[7] Aryl halides, which could be used for further transformation, also survived the reaction (entry 7). Some heterocycles such as coumarin, 2-acetylthiophene, and *N*-tosylmorpholine survived, although *N*-tosylpyrrole and 2-bromothiophene could not tolerate the reaction (entries 8−12).

As shown in Table 3, the present method can be applied to other electron-rich aromatic compounds. Methoxybenzene derivatives gave the corresponding C−N coupling products in moderate to good yields (entries 2−4). In particular, para-substituted methoxybenzenes gave single regioisomers. π-Extended aromatic compounds such as phenanthrene (**1e**) were effective in this reaction (entry 5). Furthermore, the

Table 2. Robustness Screen of C−N Coupling of *N*-Methylsulfonylimidazole with Naphthalene[a]



entry	additive	additive remaining (%)	yield of **3a** (%)	naphthalene remaining (%)
1	—	—	82	0
2		100	77	0
		90		
3		100	70	0
		98		
4		100	77	0
5		52	41	0
6		94	84	0
		90		

run	additive	additive remaining (%)	yield of **3a** (%)	naphthalene remaining (%)
7		92		
		94	88	0
		92		
8		84	76	0
9		42	39	45
10		76	27	23
11		96	88	0
12		97	77	0

[a]Yields of **3a**, the additives, and naphthalene (**1a**) after reaction were determined by GC analysis using tridecane as an internal standard. Color coding should help the ready assessment of the data: green (above 66%), yellow (34−66%), red (below 34%).

method could also be applied to other imidazole derivatives such as *N*-methylsulfonyl-2-methylimidazole (**2h**) and *N*-methylsulfonylbenzimidazole (**2i**), which gave the corresponding C−N coupling products in good yields (entries 6−7).

Benzylic compounds are also effective for the C−N coupling (entries 8−12). Notably, primary, secondary, and tertiary benzylic C−H bonds were functionalized efficiently. The regioselectivity of the reaction of 4-methoxy-1,2-dimethylbenzene (**1i**) is interesting. The para methyl group was selectively functionalized without affecting the meta methyl group (entry 11). 2-Methylimidazole and benzoimidazole could also be introduced by the present C−N coupling (entries 12 and 13).

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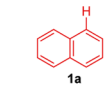
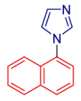
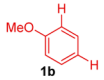
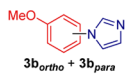
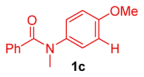
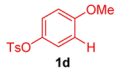
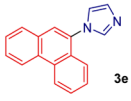
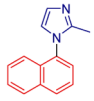
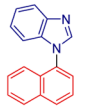
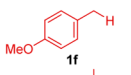
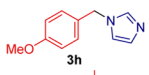
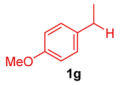
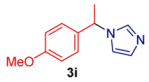
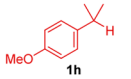
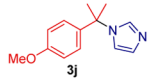
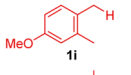
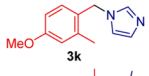
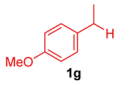
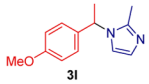
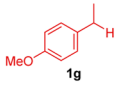
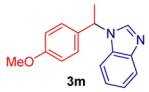
Table 3. C−N Coupling of Imidazole Derivatives with Various Aromatic and Benzylic Compounds[a]

entry	aromatic or benzylic compound	imidazole	product	yield (%)[b]
1	 1a	 2f	 3a	81
2[c]	 1b	 2f	 $3b_{ortho}$ + $3b_{para}$	68 (*o*/*p*=1/2.0)
3[c]	 1c	 2e	 3c	70
4	 1d	 2f	 3d	36
5	 1e	 2f	 3e	75
6	 1a	 2h	 3f	97
7	 1a	 2i	 3g	61
8[c]	 1f	 2f	 3h	81
9	 1g	 2f	 3i	99
10	 1h	 2f	 3j	83
11[c]	 1i	 2f	 3k	65
12	 1g	 2h	 3l	70
13	 1g	 2i	 3m	60
14[d]	 1j	2f	3n	91
15[d,e]	1k	2f	3o	77

[a]A total of 0.2 mmol of **1** was oxidized electrochemically in the presence of 0.6 mmol of N-protected imidazole **2** using 2.5 F/mol of electricity in a 1.0 M solution of $LiClO_4$ in CH_3CN at room temperature. [b]Isolated yields. [c]A total of 1.0 mmol of N-protected imidazole was used with 3.0 F/mol of electricity. The reaction was carried out at 50 °C. [d]Electrolysis was carried out in 0.1 M solution of $LiClO_4$ in 1,2-dimethoxyethane/dichloromethane (1/5). The reaction was carried out at 0 °C with 2.2 F/mol of electricity. [e]A total of 0.1 mmol of **1k** and 0.5 mmol of **2f** were used.

The present method can be integrated with electrooxidative alkene-cyclization reactions.[15] For example, the electrochemical reaction of an alkene having a nucleophilic tosylamide group in the presence of **2f** followed by the treatment with piperidine gave the cyclized *N*-benzylimidazole (entries 14 and 15). Only the five-membered ring compound was formed, and a single diastereomer was observed by ^{1}H NMR analysis of the crude product. The stereochemistry was determined by JBCA[16] and NOE analyses and is consistent with a mechanism involving the back-side attack to a benzylic cation stabilized by the cyclized tosylamide group.[10e]

The electrochemical reaction is facilitated by an electron-donating group such as a methoxy group on the aromatic ring. The use of such an activating group gives us an added bonus because a methoxy group can be used for further transformations such as transition-metal-catalyzed coupling reactions.[17] For example, compound **3i** synthesized by the present electrochemical C−N coupling was reacted with PhMgBr in the presence of a catalytic amount of $NiCl_2(PCy_3)_2$ to give compound **5**, a P450 17 inhibiter,[18] in 75% yield (Scheme 1).

Scheme 1. Synthesis of P450 17 Inhibitor 5 from 3i Featuring Nickel Catalyzed C−O Bond Activation

The following synthesis of an antifungal agent,[19] **6**, also demonstrates the power of the present method (Scheme 2).

Scheme 2. Synthesis of Antifungal Agent 6 Using Electrochemical C−N Coupling

Compound **7**, which has two different benzylic positions, was prepared from commercially available compounds in one step.[20] The present electrochemical method functionalized the benzylic C−H bond next to the electron-rich aromatic ring substituted by a methoxy group (highlighted in red) selectively without affecting the benzylic C−H bond next to the relatively electron poor aromatic ring substituted by two chloro groups (highlighted in green). Thus, the electrochemical oxidation of **7** in the presence of 1-mesylimidazole **2f** followed by treatment with piperidine gave the desired **6** in 71% isolated yield.

In conclusion, we have developed a new electrooxidative C−N coupling of *N*-mesylimidazole and its derivatives with aromatic and benzylic compounds. The key to the success of the present method is the formation of electrochemically inactive imidazolium ions as initial products, which is converted to the corresponding *N*-aryl- or *N*-benzylimidazoles by subsequent treatment with piperidine. The method provides a straightforward, highly chemoselective metal-free route to the N-substituted imidazoles.

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■ ASSOCIATED CONTENT

Supporting Information

Experimental procedures and spectroscopic data of compounds data (PDF). This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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