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Iron(III)-Catalyzed Cascade Reaction between Nitroolefins and 2-Aminopyridines: Synthesis of Imidazo[1,2-*a*]pyridines and Easy Access towards Zolimidine

Sougata Santra,[a] Avik Kumar Bagdi,[a] Adinath Majee,[a] and Alakananda Hajra[a,*]

[a] Department of Chemistry, Visva-Bharati (A Central University), Santiniketan 731235, India
Fax: (+91)-3463-261526; phone: (+91)-3463-261526; e-mail: alakananda.hajra@visva-bharati.ac.in

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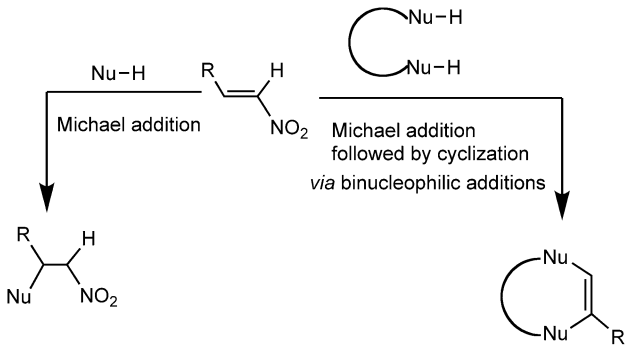
Abstract: The iron(III)-catalyzed one-pot cascade reaction between nitroolefins and 2-aminopyridines has been demonstrated for the synthesis of imidazo-[1,2-*a*]pyridines by exploiting the bielectrophilic nature of nitroolefins. This methodology could be successfully applicable for the synthesis of zolimidine, a useful drug for the treatment of peptic ulcer. The reaction proceeds through Michael addition followed by intramolecular cyclization and *in situ* denitration.

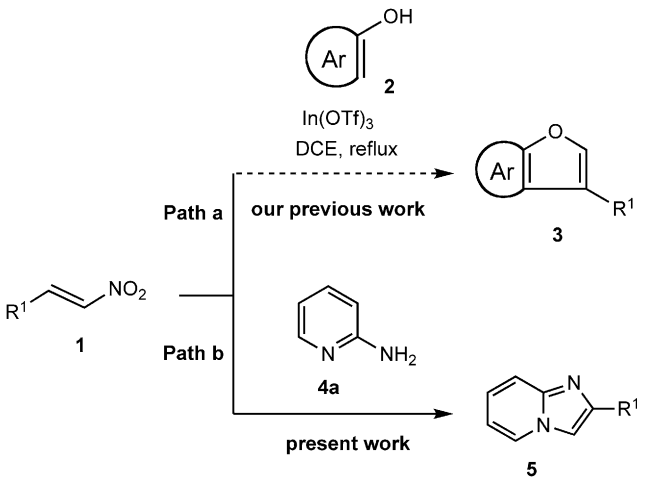
Keywords: cascade reactions; imidazo[1,2-*a*]pyridines; iron(III) chloride; nitroolefins; zolimidine



Scheme 1. Various modes of addition of nucleophiles to nitroalkenes.

Cascade reactions are one of the most powerful and atom economical methodologies in contemporary organic synthesis.[1] These reactions usually provide processes in a more efficient and environmentally benign manner than conventional procedures by omitting the steps of separation and purification of the reaction intermediates. The conjugate addition of nucleophiles to α,β-unsaturated compounds (such as α,β-unsaturated carbonyl compounds, esters, nitriles, phosphates, sulfones, and nitroalkenes) is known as the Michael addition.[2] It is a versatile reaction for carbon-carbon and carbon-heteroatom bond formations. Both Michael and hetero-Michael additions are widely used in inter- and intramolecular reactions to provide biologically important scaffolds. Among the various commonly used Michael acceptors nitroolefins are excellent substrates for 1,4-addition due to the strong electron-withdrawing ability of the nitro group and low tendency for 1,2-addition.[3] In addition nitroalkenes are bielectrophilic in nature and they are potentially useful for multiple nucleophilic additions to form heterocycles in a cascade fashion (Scheme 1). Although various methodologies have been developed for the 1,4-conjugate addition, less attention has been paid to investigate the bielectrophilic nature of nitroalkenes and their applications in organic synthesis to afford the carbocyclic and heterocyclic compounds in a systematic way.

Recently, we have described a simple, and straightforward one-pot synthesis of benzofuran and naphthofuran derivatives from readily available α,β-unsaturated nitroalkenes by coupling with phenols/naphthols. (Scheme 2, Path a).[4] A Michael addition of phenol/naphthols to α,β-unsaturated nitroalkenes and followed by intramolecular Friedel–Crafts cyclization are the key steps for this methodology and are followed by *in situ* denitration process to give the product. Encouraged by this result, we continued our interest to explore this methodology for the synthesis of various important building blocks. Consequently, we searched for a precursor having two nucleophilic sites capable of double nucleophilic addition to nitroalkenes. We chose 2-aminopyridine as it contains two nucleophilic nitrogen atoms which might be capable for multiple nucleophilic additions to the nitroalkenes to produce imidazopyridine derivatives (Scheme 2, Path b).



Scheme 2. Cascade reaction of nitroolefins with binucleophiles.

The imidazopyridine nucleus, in particular the imidazo[1,2-*a*]pyridine core, represents a significant class of biologically active nitrogen compounds that exhibit a number of important biological properties,[5] such as antifungal,[6] anti-inflammatory,[7] antitumor,[8] antiviral,[9] antibacterial,[10] antiprotozoal,[11] antipyretic,[12] analgesic,[13] antiapoptotic,[14] hypnoselective, and anxioselective activities.[15] Recently, imidazo[1,2-*a*]pyridine scaffolds were incorporated in some commercially available drugs[16] such as zolpidem (**I**, used in the treatment of insomnia), alpidem (**II**, as an anxiolytic agent), olprinone (**III**, for the treatment of acute heart failure), zolimidine (**IV**, used for the treatment of peptic ulcer), necopidem and saripidem (**V** and **VI**, both work as an anxiolytic agent) (Figure 1).

Considering these important uses, their syntheses have been received much attention in the field of medicinal and pharmaceutical chemistry. Several methods are available for the synthesis of imidazo[1,2-*a*]pyridines such as condensation of 2-aminopyridine with α-halo carbonyl compounds,[17] one-pot condensations of aldehydes, isonitriles, and 2-aminopyridines,[18] copper-catalyzed three-component reactions of 2-aminopyridines, aldehydes, and alkynes,[19] oxidative couplings through C–H activation,[20] and very recently, from Morita–Baylis–Hillman acetates of nitroalkenes.[21] However, most of these methodologies have been developed using expensive reagents, commercially less available alkynes, and α-halogeno carbonyl compounds which have lachrimatory properties. Therefore, finding a new methodology for the synthesis of imidazo[1,2-*a*]pyridines in terms of efficiency, operational simplicity, availability of starting materials, and economic practicability is highly desirable. Over the past few years iron salts have been shown to be effective alternatives and promising transition metal catalysts and have received much attention due to their low price, sustainability, ready availability, non-toxicity, and environmentally friendly properties.[22] In continuation of our current research[4] to develop newer methodologies using less toxic and inexpensive metals as a catalyst,[23] herein, we are pleased to report a convenient approach to access imidazo[1,2-*a*]pyridines through iron(III)-catalyzed binucleophilic addition of 2-aminopyridines to nitroalkenes (Scheme 3).

Optimization of the reaction conditions was achieved by varying temperature, solvent, and catalyst (Table 1). For the initial study, nitrostyrene **1a** and 2-aminopyridine **4a** were selected as the model substrates to optimize the reaction conditions. Initially we carried out the reaction in the presence of a catalytic amount of indium triflate (5 mol%) in 1,2-dichloroethane under reflux. Unfortunately, no desired

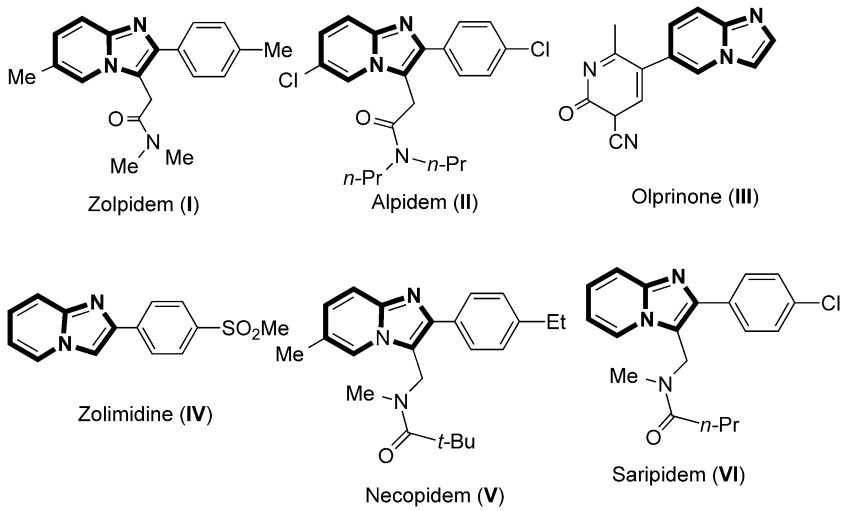
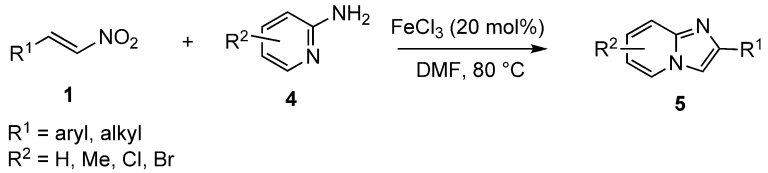
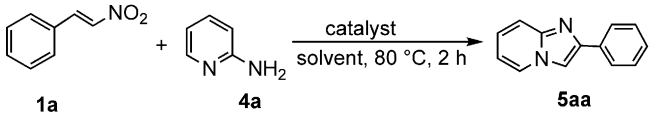


Figure 1. Imidazo[1,2-*a*]pyridine-containing drugs.



Scheme 3. Synthesis of imidazo[1,2-*a*]pyridines.

Table 1. Optimization of reaction conditions.[a]



1a **4a** **5aa**

Entry	Catalyst (mol%)	Solvent	Yield of **5aa** [%][b]
1[c]	$In(OTf)_3$ (5 mol%)	DCE	0
2	$In(OTf)_3$ (5 mol%)	DMF	<10
3	$In(OTf)_3$ (10 mol%)	DMF	<10
4[d]	CuI (5 mol%)	DMF	16
5[d]	$Cu(OAc)_2$ (5 mol%)	DMF	14
6[d]	$Cu(OTf)_2$ (10 mol%)	DMF	15
7	$FeCl_3$ (10 mol%)	DMF	72
8	$FeBr_3$ (10 mol%)	DMF	45
9	$Fe(OTf)_3$ (10 mol%)	DMF	30
10	$AlCl_3$ (10 mol%)	DMF	<5
11	$ZnCl_2$ (10 mol%)	DMF	<5
12	$LaCl_3$ (10 mol%)	DMF	<5
13	$BF_3{\cdot}Et_2O$ (10 mol%)	DMF	<5
14	**$FeCl_3$ (20 mol%)**	**DMF**	**84**
15	$FeCl_3$ (30 mol%)	DMF	85
16	$FeCl_3$ (20 mol%)	DMSO	48
17	$FeCl_3$ (20 mol%)	CH_3CN	37
18[e]	$FeCl_3$ (20 mol%)	H_2O	18
19	–	DMF	NR[f]

[a] Carried out with 1 mmol of **1a** and 1 mmol of **4a** in the presence of catalyst in solvent (2 mL) at 80 °C for 2 h.
[b] Isolated yields.
[c] Reaction proceeded under reflux.
[d] Major product was obtained through oxidative coupling (Ref.[24]).
[e] Reaction carried out at 100 °C.
[f] No formation of **5aa**.

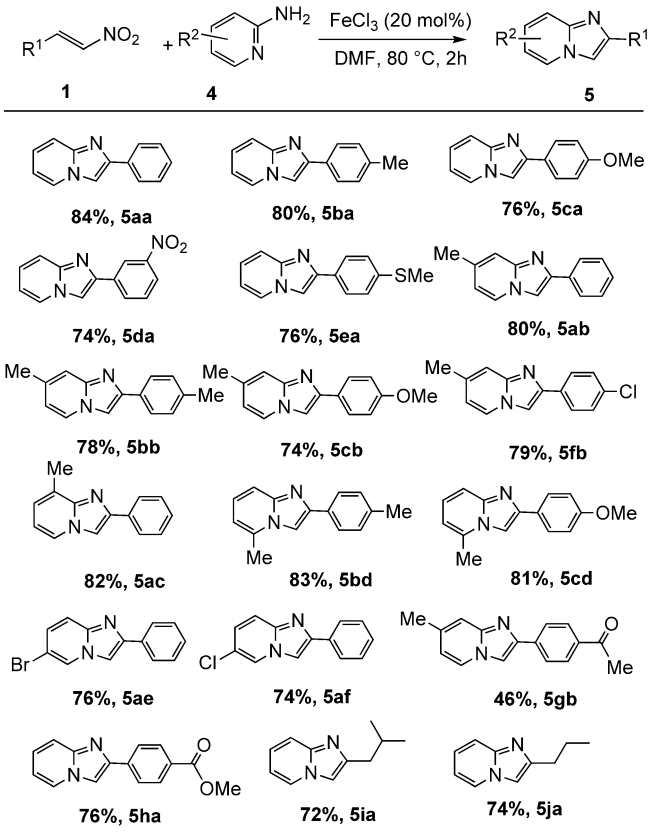
product was obtained. Next we changed the solvent to DMF and the product was formed in less than 10%. Encouraged by this result, we tried the reaction using Cu salts as catalyst. However, a different product was obtained in addition to the desired product in 14–16%. The undesired product is 3-nitro-2-phenylimidazo[1,2-*a*]pyridine and probably its formation occurred through the oxidative coupling of a nitroolefin with 2-aminopyridine.[24] Next, we shifted to iron salts as catalyst. $FeCl_3$ was found to be the most effective one among various iron salts such as $FeCl_3$, $FeBr_3$, $Fe(OTf)_3$. However, other common Lewis acids were not effective for this conversion (entries 10–13, Table 1). Finally, the optimized condition were obtained using 20 mol% $FeCl_3$ at 80 °C in DMF as solvent. DMF appeared to be the best choice among common solvents such as H_2O, DMSO, CH_3CN. In absence of catalyst, no formation of the desired product was observed. 20 mol% was required as optimum amount of catalyst and increasing the amount of catalyst did not improve the yields while decreasing the amount of catalyst decreased the yields.

With the optimized conditions in hand, we then turned our attention to investigate the scope of substrates, and the results are shown in Table 2. A wide range of both substituted aminopyridines and nitroolefins were employed to prove the general applicability of our present procedure. Several sensitive functionalities such as, OMe (**5ca**, **5cb**, **5cd**), NO_2 (**5da**), halogens Cl, Br (**5ae**, **5af**, **5fb**), COMe (**5gb**), CO_2Me (**5ha**) were unaffected under the present reaction conditions. Substituted 2-aminopyridines with the methyl group at different positions reacted with nitroolefins in good yields (**5ab**, **5bb**, **5cb**, **5fb**, **5ac**, **5bd**, **5cd**, **5gb**). In addition, aliphatic nitroalkenes were also found to afford the desired products with high yields (**5ia**, **5ja**). However, β-methyl-β-nitrostyrene did not give the desired heterocycles under the present reaction conditions.

The SMe group was also well tolerated under these reaction conditions. We have successfully synthesized the corresponding compound **5ea** with high yield. It is noteworthy to mention that the compound **5ea** is the key intermediate for synthesizing the drug zolimidine. After successive oxidation of **5ea** using *m*CPBA, the target molecule can be synthesized by employing the reported method (Scheme 4).[20j]

A library of 3-unsubstituted imidazo[1,2-*a*]pyridine derivatives was synthesized employing the present method. Further functionalization was carried out at the 3-position to construct densely substituted imidazo[1,2-*a*]pyridine derivatives by modified meth-

Table 2. Substrate scope for the reaction of nitroolefins **1** and 2-aminopyridines **4**.[a]

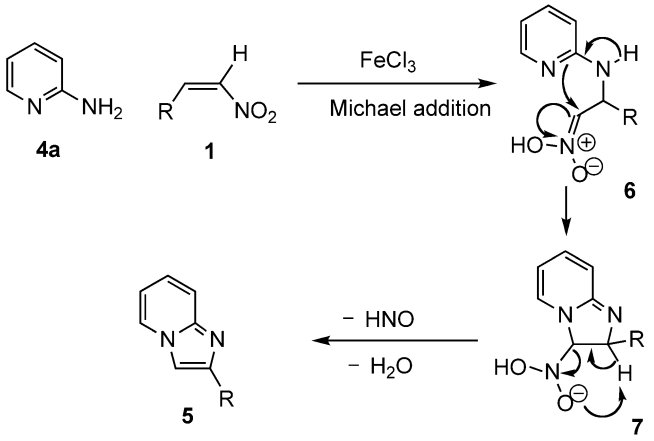


[a] *Reactions conditions:* 1.0 mmol of **1** and 1.2 mmol of **4** in the presence of $FeCl_3$ (20 mol%) in 2 mL of DMF at 80 °C for 2 h, isolated yields.

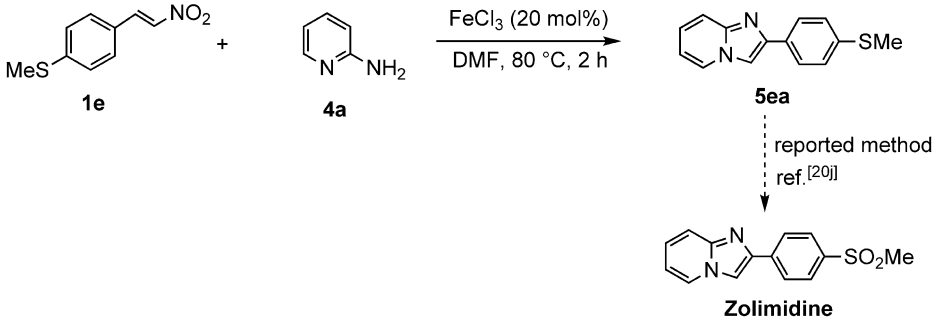
ods (Scheme 5).[25] 3-Aryl-substituted imidazo[1,2-*a*]pyridines were obtained in high yields.

A plausible mechanism for this iron(III)-catalyzed cascade reaction is outlined in Scheme 6. As described previously[4] the first step of the reaction is the Michael addition of 2-aminopyridine **4a** with nitroolefin **1** to form the intermediate **6**. Probably Fe(III) chloride accelerated the reaction by increasing the electrophilicity of the nitroolefin through coordination. The second step is the intramolecular cyclization of the intermediate **6** leading to the final product after subsequent removal of water and nitroxyl (HNO).[26]

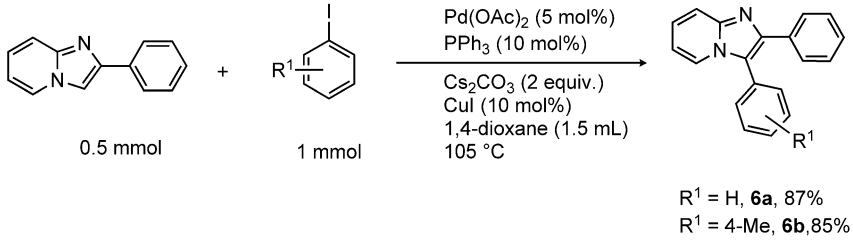
In conclusion, we have developed an iron(III)-catalyzed one-pot tandem reaction for the synthesis of



Scheme 6. A plausible reaction mechanism.



Scheme 4. Synthetic approach towards zolimidine.



Scheme 5. Functionalization of 3-unsubstituted imidazopyridines.

imidazo[1,2-*a*]pyridines by exploiting the bielectrophilic nature of nitroalkenes and the binucleophilic nature of 2-aminopyridines. This methodology could be successfully applicable for the synthesis of zolimidine, a useful drug for the treatment of peptic ulcer. Easily available starting materials, as well as the non-hazardous and less expensive catalyst are the advantages of the present procedure. Further study to explore the bielectrophilic nature of nitroalkenes to construct the biologically active molecules is ongoing in our laboratory.

Experimental Section

Typical Procedure for the Synthesis of 2-Phenylimidazo[1,2-*a*]pyridine (5aa)

A mixture of 2-aminopyridine **4a** (112 mg, 1.2 mmol) and nitroalkene **1a** (149 mg, 1 mmol) was stirred in presence of anhydrous $FeCl_3$ (20 mol%) in DMF (2 mL) at 80 °C for 2 h (TLC). After completion, the reaction mixture was cooled to room temperature and extracted with dichloromethane (10 mL) followed by washing with brine (5 mL) and drying over Na_2SO_4. After evaporation of the solvent the crude product was purified by column chromatography on silica gel using petroleum ether/ethyl acetate (3:1 to 2:1) as eluent.

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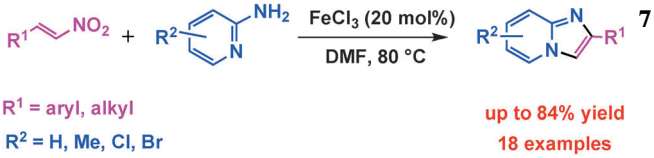
Sougata Santra, Avik Kumar Bagdi, Adinath Majee, Alakananda Hajra*



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