EurJOC European Journal of Organic Chemistry

FULL PAPER

DOI: 10.1002/ejoc.201301658

# Iron(III)-Catalyzed Denitration Reaction: One-Pot Three-Component Synthesis of Imidazo[1,2-*a*]pyridine Derivatives

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**Keywords:** Synthetic methods / Multicomponent reactions / Nitrogen heterocycles / Iron

An iron(III)-catalyzed one-pot three-component cross-coupling nitration reaction of 2-aminopyridines, aldehydes and nitroalkanes, straightforwardly forms imidazo[1,2-*a*]pyridine derivatives and is described in this report. The system shows good functional-group tolerance and proceeds smoothly in moderate to good yields.

## Introduction

Multicomponent reactions[1] have been used frequently as powerful methods in material, medical, drug, and organic chemistry to provide structural diversity[2] by synthetic chemists. Various multicomponent reactions have proven to be efficient and have received considerable interest because they offer prominent advantages such as reducing time, saving cost and energy, and also create complex structures from commercially available starting materials. Considering the features of multicomponent reactions, developing new and efficient routes for their synthesis has been recognized as one of the most important topics in recent years.

The imidazo[1,2-*a*]pyridine scaffold is a 5,6-fused nitrogen bridgehead heterocycle, which is one of the most significant structural units. It exhibits widespread applications in medicinal chemistry[3] and materials science,[4] and is also found in important pharmaceuticals and various bioactive compounds (Figure 1), such as saripidem (**I**),[5] olprinone (**II**),[6] zolimidine (**III**),[7] zolpidem(**IV**),[8] soraprazan (**V**),[9] and alpidem (**VI**).[10] Consequently, the construction of imidazo[1,2-*a*]pyridines has attracted a considerable attention.[11] Recently, with the development of transition-metal-catalyzed cascade reactions,[12] a variety of approaches based on N–H and C–H activation have been discovered.[13] Gryko and co-workers reported the condensation of 2-aminopyridines with α-halogeno ketones to synthesize imidazo[1,2-*a*]pyridines.[14] Construction of imidazo[1,2-*a*]-pyridines through three-component coupling of 2-aminopyridines, aldehydes and isonitriles[13b,15] was achieved by the DiMauro group and Adib group, respectively. Namboothiri et al. have synthesized imidazo[1,2-*a*]pyridines from Morita–Baylis–Hillman acetates of nitroalkenes,[11] and we obtained imidazo[1,2-*a*]pyridines by using 2-aminopyridine and 2-methyl-nitroolefins.[16] Although the literature on imidazo[1,2-*a*]pyridine syntheses has enjoyed a rich history of versatile methods, most of these methods are not environmentally benign owing to the use of halogeno compounds, or are limited because of multistep preparations of the starting materials. Therefore, finding an easy and efficient method to construct imidazo[1,2-*a*]pyridines by using environmentally friendly reagents with simple and readily accessible substrates is still desirable. Iron is one of the most abundant, inexpensive and environmentally friendly metals[17] on earth. In addition, many iron salts and complexes are readily accessible, thus, iron is highly valued for easy and efficient methodologies to molecular construction. To the best of our knowledge, only a few approaches have been reported based on iron-catalyzed denitration[18] reactions. For example, Chen and co-workers introduced the construction of imidazole derivatives by using this strategy. The Hajra group also reported the synthesis of imidazo[1,2-*a*]pyridines from 2-aminopyridine and nitroolefins through iron-catalyzed denitration reaction.[18a] In spite of these methods, synthesizing imidazo[1,2-*a*]pyridines through an iron-catalyzed denitration multicomponent reaction has rarely been reported. As a part of our continuing studies into the synthesis of heterocycles,[19] we paid attention to designing a new strategy for straightforward access to imidazo[1,2-*a*]pyridine rings through a denitration reaction catalyzed by iron(III) in a one-pot three-component reaction, involving the assembly of the scaffold from [3+1+1] atom fragments (Scheme 1). The method is facile,

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Supporting information for this article is available on the WWW under http://dx.doi.org/10.1002/ejoc.201301658.

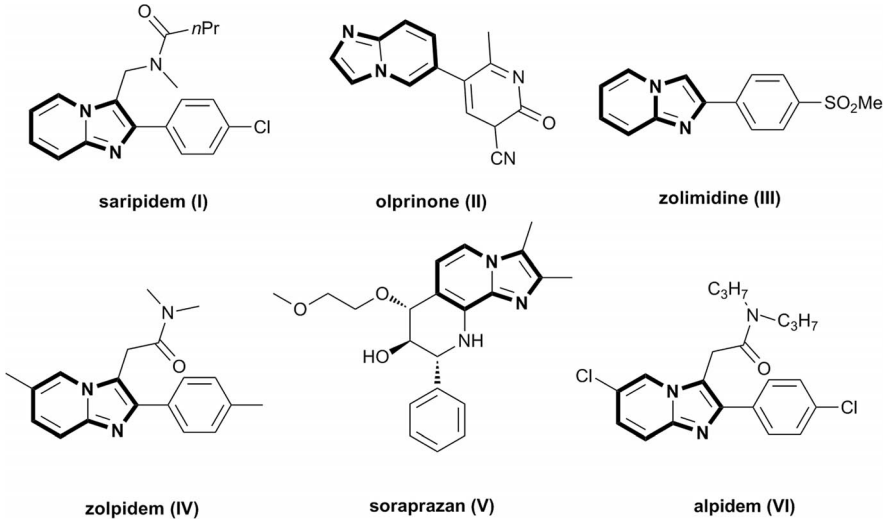
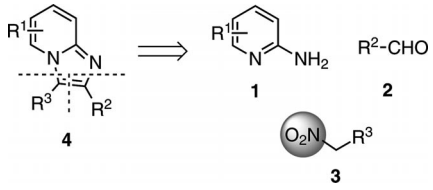



Figure 1. Examples of imidazo[1,2-*a*]pyridines with biological activities.

simple to undertake, uses commercially available starting materials, is environmentally benign, and shows functional group tolerance. Herein, we wish to present this simple one-pot synthesis of imidazo[1,2-*a*]pyridine derivatives.

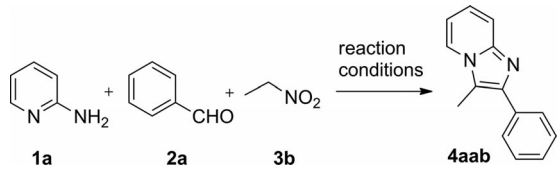


Scheme 1. Synthesis of imidazo[1,2-*a*]pyridines thorugh a [3+1+1] reaction.

## Results and Discussion

Initially, the three-component reaction of 2-aminopyridine (**1a**), benzaldehyde (**2a**) and $EtNO_2$ (**3b**) was chosen as the model. The results are summarized in Table 1. When the reaction was performed by using $FeCl_2$ as the catalyst at 70 °C for 4 hours in air, desired 3-methyl-2-arylimidazo[1,2-*a*]pyridine (**4aab**) was obtained in 53% yield (Table 1, Entry 1). Then different iron salts were evaluated. The screening of iron catalysts showed $FeCl_3$ to be superior relative to $FeCl_2$, $FeSO_4$, $Fe(acac)_3$, $FeSO_4{\cdot}7H_2O$, $FeCl_3$ and $AgNO_3$ (Table 1, Entries 1–6). In addition, no product was observed without the presence of metal iron salts (Table 1, Entry 11). Carrying out the reaction at 90 °C improved the yield (Table 1, Entry 7), whereas a continued rise in temperature did not significantly ameliorate the yield (Table 1, Entry 8). A longer or shorter reaction time did not promote the transformation (Table 1, Entries 9 and 10). Importantly, when the amount of $EtNO_2$ was reduced to 10 equivalents, the product yield did not decrease proportionately (Table 1, Entry 13); however, only 60% yield (**4aab**) was isolated in the presence of 5 equiv. of $EtNO_2$ (Table 1, Entry 12). Furthermore, when the reaction was conducted in either $O_2$ or $N_2$, lower product yields were obtained (Table 1, Entry 14 and 15). Finally, the optimized conditions were identified (Table 1, Entry 13).

Table 1. Optimization of reaction conditions.[a]




| Entry | Catalyst | *t* [h] | *T* [°C] | Yield [%][b] |
|---|---|---|---|---|
| 1 | $FeCl_2$ | 4 | 70 | 53 |
| 2 | $FeCl_3$ | 4 | 70 | 58 |
| 3 | $FeSO_4$ | 4 | 70 | 55 |
| 4 | $Fe(acac)_3$ | 4 | 70 | 0 |
| 5 | $FeSO_4{\cdot}7H_2O$ | 4 | 70 | 50 |
| 6 | $FeCl_3$ and $AgNO_3$ | 4 | 70 | trace[c] |
| 7 | $FeCl_3$ | 4 | 90 | 75 |
| 8 | $FeCl_3$ | 4 | 110 | 75 |
| 9 | $FeCl_3$ | 2 | 90 | 73 |
| 10 | $FeCl_3$ | 6 | 90 | 73 |
| 11 | – | 4 | 90 | 0 |
| 12 | $FeCl_3$ | 4 | 90 | 60[d] |
| 13 | $FeCl_3$ | 4 | 90 | 75[e] |
| 14 | $FeCl_3$ | 4 | 90 | 56[f] |
| 15 | $FeCl_3$ | 4 | 90 | 61[g] |

[a] The reaction was carried out with **1a** (0.5 mmol), **2a** (0.6 mmol), and **3b** (15 equiv.) with catalyst (0.05 mmol) under air. [b] Yield of isolated product. [c] $FeCl_3$ (0.05 mmol) and $AgNO_3$ (0.05 mmol) were used. [d] 5 equiv. of **3b** were used. [e] 10 equiv. of **3b** were used. [f] The reaction was performed under an $O_2$ atmosphere. [g] The reaction was performed under a $N_2$ atmosphere.

Having established the optimal reaction conditions, the scope with substituted aminopyridines was investigated. The results are illustrated in Table 2 and show that a variety of substituted aminopyridines with benzaldehyde and $EtNO_2$ mostly proceed smoothly and to give the corresponding products in moderate to good yields. Various elec-

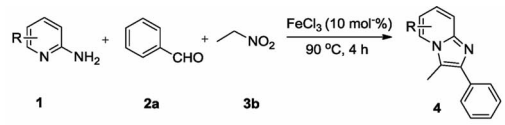
tron-donating (Me, OBn; Table 2, Entries 1–4, and 10) and electron-withdrawing substituents (Cl, Br, F, COOEt; Table 2, Entries 5–9) on the aminopyridines were tolerated in this transformation. After further investigation, we found that the nature of the substituents on the aminopyridines had some influence on the product yields. 2-Aminopyridines substituted with electron-deficient groups (Table 2, Entries 5–9) resulted in higher yields than those with electron-rich ones (Table 2, Entries 1–4, and 10). 5-Substituted 2-aminopyridines gave products in highest yield (Table 2, Entries 6 and 8). Furthermore, several 2-amino heterocycles, such as 2-aminopyrimidine and 2-aminobenzimidazole, were unsuitable for the process and the desired products were not isolated, and only a trace amount of product was detected from TLC measurement.

Next, the substrate scope with aldehydes was further examined, as shown in Table 3. We were pleased to note that the transformation is very general for a wide range of aldehydes. Aryl and alkyl aldehydes revealed good reactivity in this reaction. A variety of electron-donating (Me, OMe; Table 3, Entries 2–6) and electron-withdrawing substituents (Cl, Br, F, CN; Table 3, Entries 7–13, 16 and 17) at the aromatic ring of the aldehyde were tolerated. In addition, a bulky naphthyl substituent did not hamper the process and also gave desired product **4anb**, albeit in lower yields (Table 3, Entry 14). Heteroaryl-substituted substrate furan-2-carbaldehyde could be converted into target product **4aob** in 57% yield (Table 3, Entry 15). Aliphatic aldehyde (3,7-dimethyloct-6-en-1-al) was less reactive and led to compound **4arb** in 40% yield (Table 3, Entry 18).

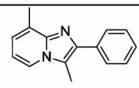
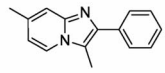
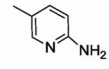
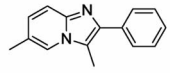
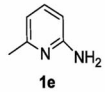
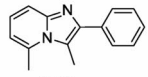
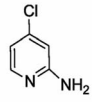
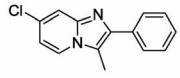
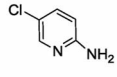
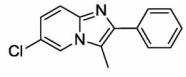
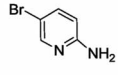
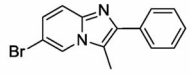
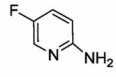
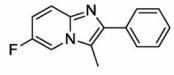
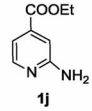
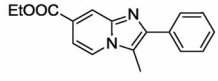
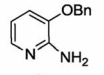
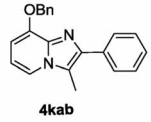
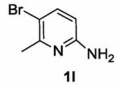
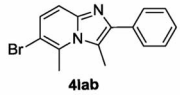
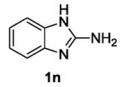
Encouraged by the results, we explored the applicability of this three-component coupling reaction. Under slightly modified reaction conditions with $FeCl_3$ as catalyst and tetrabutylammonium iodide (TBAI) as the base, which could improve the reaction yield, and $MeNO_2$ as substrate, desired 3-unsubstituted imidazopyridine **4aaa** was obtained in good yield (Table 4, Entry 1). Then we evaluated the scope of the reaction. As depicted in Table 4, the reaction proceeded smoothly, but the nature of substituents on the aryl ring affected the reaction. Substitutes with electron-deficient groups (Table 4, Entries 4–6) resulted in higher yields than those with electron-rich ones (Table 4, Entries 2, 3, and 7). Moreover, the SMe group was also well tolerated. Notably, the reaction with 4-(methylthio) benzaldehyde gave product **4asa** in moderate yield. Further oxidation of **4asa** by using *meta*-chloroperoxybenzoic acid could synthesize the drug zolimidine according to the literature (Scheme 2).[20]

To further demonstrate the use of this method, 2-aminopyridines, benzaldehyde and 1-nitropropane were employed as substrates under standard reaction conditions. The reaction proceeded smoothly, and expected 3-ethyl-2-phenylimidazo[1,2-*a*]pyridine (**4aac**) was obtained in 75% yield (Table 5, Entry 1). Next, we explored the variation of the benzaldehyde in reactions with 2-aminopyridines, and 1-nitropropane. As expected, the substituent effect was consistent with $MeNO_2$ and $EtNO_2$. Substituents with electron-donating group (Table 5, Entries 2–4) resulted in lower

Table 2. Substrates scope with aminopyridines.[a]



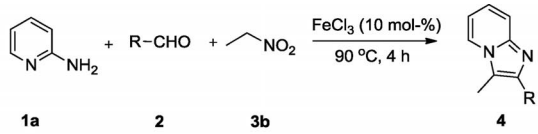

1 2a 3b 4

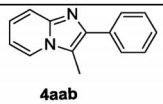
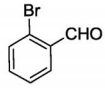
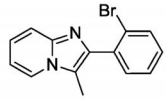
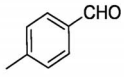
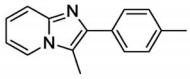
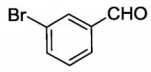
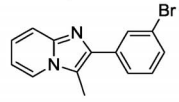
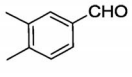
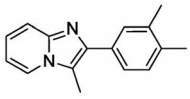
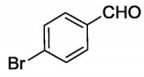
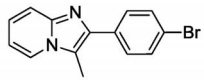
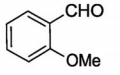
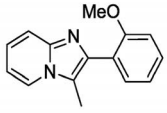
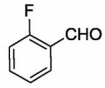
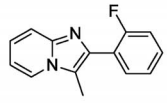
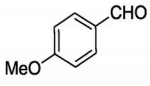
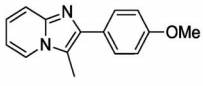
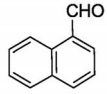
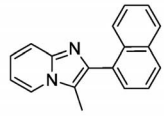
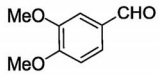
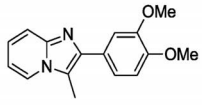
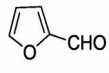
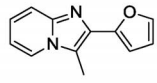
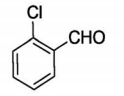
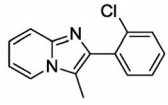
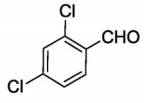
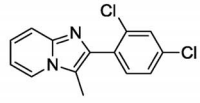
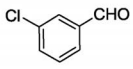
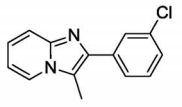
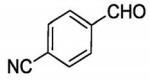
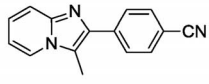
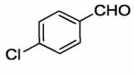
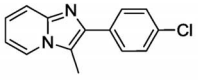
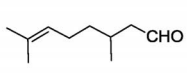
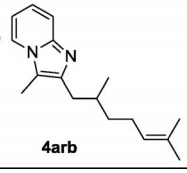
| Entry | Aminopyridine | Product | Yield [%][b] |
|---|---|---|---|
| 1 |  1b |  4bab | 50 |
| 2 |  1c |  4cab | 66 |
| 3 |  1d |  4dab | 64 |
| 4 |  1e |  4eab | 35 |
| 5 |  1f |  4fab | 73 |
| 6 |  1g |  4gab | 85 |
| 7 |  1h |  4hab | 72 |
| 8 |  1i |  4iab | 78 |
| 9 |  1j |  4jab | 76 |
| 10 |  1k |  4kab | 55 |
| 11 |  1l |  4lab | 62 |
| 12 |  1m | not isolated | trace |
| 13 |  1n | not isolated | trace |

[a] The reaction was carried out by using **1** (0.5 mmol), **2a** (0.6 mmol), and **3b** (10 equiv.) with catalyst (0.05 mmol) in air. [b] Yield of isolated product.

Table 3. Substrate scope with aldehydes.




| Entry | Aldehyde | Product | Yield [%][b] | Entry | Aldehyde | Product | Yield [%][b] |
|---|---|---|---|---|---|---|---|
| 1 |  **2a** |  **4aab** | 75 | 10 |  **2j** |  **4ajb** | 60 |
| 2 |  **2b** |  **4abb** | 46 | 11 |  **2k** |  **4akb** | 68 |
| 3 |  **2c** |  **4acb** | 56 | 12 |  **2l** |  **4alb** | 66 |
| 4 |  **2d** |  **4adb** | 66 | 13 |  **2m** |  **4amb** | 87 |
| 5 |  **2e** |  **4aeb** | 57 | 14 |  **2n** |  **4anb** | 40 |
| 6 |  **2f** |  **4afb** | 40 | 15 |  **2o** |  **4aob** | 57 |
| 7 |  **2g** |  **4agb** | 56 | 16 |  **2p** |  **4apb** | 65 |
| 8 |  **2h** |  **4ahb** | 23 | 17 |  **2q** |  **4aqb** | 50 |
| 9 |  **2i** |  **4aib** | 50 | 18 |  **2r** |  **4arb** | 40 |

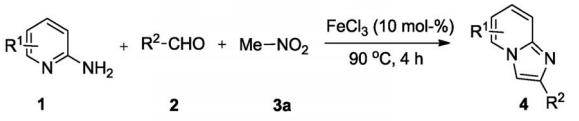
[a] The reaction was carried out by using **1a** (0.5 mmol), **2** (0.6 mmol), and **3b** (10 equiv.) with catalyst (0.05 mmol) in air. [b] Yield of isolated product.

yields than those with electron-withdrawing groups (Table 5, Entries 6–9).

On the basis of the above experimental observations, a plausible mechanism for this reaction is outlined in Scheme 3. The first step involves coupling of substrates **1** and **2** to produce imine **5**. Presumably, $Fe^{III}$ chloride acts as a Lewis acid that promotes the reaction by increasing the electrophilicity of imine **5** through coordination, providing a good electrophilic substrate for nucleophilic nitroalkane **3**. Then **3** reacts with imine **5** through a Michael addition reaction to give intermediate **6**. Subsequently, an internal proton transfer and intramolecular second Michael

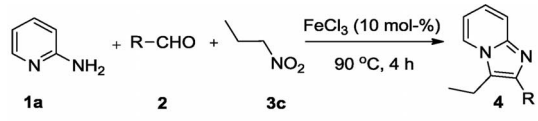
Table 4. Substrate scope with aminopyridines and aldehydes.




| Entry | Amino-pyridine | Aldehyde | Product | Yield [%][b] |
|---|---|---|---|---|
| 1 | 1a | 2a | 4aaa | 70 |
| 2 | 1a | 2b | 4aba | 30 |
| 3 | 1a | 2d | 4ada | 35 |
| 4 | 1a | 2k | 4aka | 55 |
| 5 | 1a | 2m | 4ama | 60 |
| 6 | 1a | 2q | 4aqa | 48 |
| 7 | 1b | 2a | 4baa | 33 |

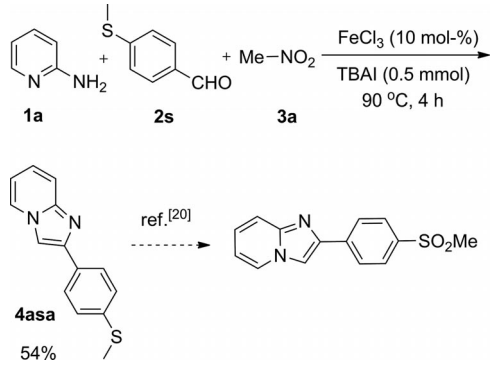
[a] The reaction was carried out by using **1** (0.5 mmol), **2** (0.6 mmol), TBAI (0.5 mmol) and **3a** (10 equiv.) with catalyst (0.05 mmol) in air. [b] Yield of isolated product.

Table 5. Substrates scope of aldehydes.



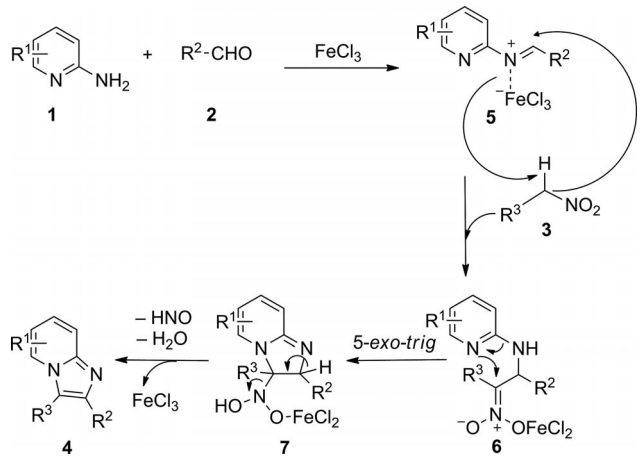

| Entry | Aldehyde | Product | Yield [%][b] |
|---|---|---|---|
| 1 | 2a | 4aac | 75 |
| 2 | 2b | 4abc | 35 |
| 3 | 2e | 4aec | 36 |
| 5 | 2f | 4afc | 30 |
| 6 | 2i | 4aic | 45 |
| 7 | 2j | 4ajc | 60 |
| 8 | 2m | 4amc | 62 |
| 9 | 2q | 4aqc | 50 |

[a] The reaction was carried out by using **1a** (0.5 mmol), **2** (0.6 mmol), and **3c** (10 equiv.) with catalyst (0.05 mmol) in air. [b] Yield of isolated product.




Scheme 2. Synthetic approach towards zolimidine.

addition reaction from the pyridine nitrogen in a regioselective 5-*exo-trig* fashion lead to intermediate **7**. Finally, intermediate **7**, upon the removal of water and nitroxyl and the release of the catalyst, collapses to product **4**.[21]




Scheme 3. Plausible reaction mechanism.

## Conclusions

In conclusion, we have successfully developed an iron(III)-catalyzed one-pot three-component cross-coupling nitration reaction of 2-aminopyridines, aldehydes, and nitroalkane, giving rise to the straightforward formation of imidazo[1,2-*a*]pyridine derivatives. Substrates that are commercially available and an inexpensive and nonhazardous catalyst are the highlights of this process. Various functional groups are tolerated and proceed smoothly in moderate to good yields.

## Experimental Section

**General Remarks:** Column chromatography was carried out with silica gel. $^1$H NMR spectra were recorded at 400 MHz in $CDCl_3$ and $^{13}$C NMR spectra were recorded at 100 MHz in $CDCl_3$ by using TMS as the internal standard. All new compounds were further characterized by HRMS; copies of their $^1$H NMR and $^{13}$C NMR spectra are provided. Commercially available reagents and solvents were used without further purification.

**Typical Procedure for the Preparation of 3-Methyl-2-arylimidazo[1,2-*a*]pyridine Derivatives:** A test tube was charged with **1a** (0.5 mmol), **2a** (0.6 mmol) and $FeCl_3$ (8.13 mg, 0.05 mmol). Then **3b** (10 equiv.) was added to the reaction system. The reaction was stirred at 90 °C under air for 4 h. After cooling to room temperature, the solvent diluted with ethyl acetate (10 mL) was evaporated in vacuo, and the residues were purified by column chromatography (petroleum ether/EtOAc) to afford pure **4aab**.

**Characterization Data of 3-Methyl-2-arylimidazo[1,2-*a*]pyridine Derivatives 4**

**3-Methyl-2-phenylimidazo[1,2-*a*]pyridine (4aab):** White solid, m.p. 144–146 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.84 (d, $J$ = 6.8 Hz, 1 H), 7.79 (d, $J$ = 7.2 Hz, 2 H), 7.62 (d, $J$ = 8.8 Hz, 1 H), 7.47–7.44 (m, 2 H), 7.35–7.32 (m, 1 H), 7.16–7.12 (m, 1 H), 6.81–6.78 (m, 1 H), 2.59 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.2, 142.3, 134.8, 128.4, 128.2, 127.2, 123.4, 122.7, 117.3, 115.8, 111.9, 9.5 ppm. HRMS (ESI): calcd. for $C_{14}H_{13}N_2$ [M + H]$^+$ 209.1073; found 209.1075.

**3-Methyl-2-(*p*-tolyl)imidazo[1,2-*a*]pyridine (4abb):** Yellow viscous liquid. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.88 (d, $J$ = 6.8 Hz, 1 H), 7.69 (d, $J$ = 8.4 Hz, 2 H), 7.63 (d, $J$ = 9.2 Hz, 1 H), 7.28 (d, $J$ = 8.0 Hz, 2 H), 7.18–7.14 (m, 1 H), 6.83 (td, $J$ = 6.8, $J$ = 1.2 Hz, 1 H), 2.63 (s, 3 H), 2.41 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.3, 142.5, 137.1, 132.0, 129.2, 128.2, 123.3, 122.8, 117.4, 115.6, 111.9, 21.2, 9.6 ppm. HRMS (ESI): calcd. for $C_{15}H_{15}N_2$ [M + H]$^+$ 223.1230; found 223.1236.

**2-(3,4-Dimethylphenyl)-3-methylimidazo[1,2-*a*]pyridine (4acb):** Yellow solid, m.p. 103–105 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.83 (d, $J$ = 6.8 Hz, 1 H), 7.62 (d, $J$ = 9.2 Hz, 2 H), 7.49 (dd, $J$ = 7.6, $J$ = 1.6 Hz, 1 H), 7.21 (d, $J$ = 8.0 Hz, 1 H), 7.15–7.10 (m, 1 H), 6.80–6.76 (m, 1 H), 2.59 (s, 3 H), 2.33 (s, 3 H), 2.30 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.1, 142.4, 136.7, 135.7, 132.3, 129.6, 129.5, 125.5, 123.2, 122.6, 117.2, 115.4, 111.8, 19.7, 19.5, 9.5 ppm. HRMS (ESI): calcd. for $C_{16}H_{17}N_2$ [M + H]$^+$ 237.1386; found 237.1383.

**2-(2-Methoxyphenyl)-3-methylimidazo[1,2-*a*]pyridine (4adb):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.86 (d, $J$ = 6.8 Hz, 1 H), 7.62 (d, $J$ = 9.2 Hz, 1 H), 7.59–7.56 (m, 1 H), 7.38–7.34 (m, 1 H), 7.14–7.10 (m, 1 H), 7.08–7.04 (m, 1 H), 6.99 (d, $J$ = 8.4 Hz, 1 H), 6.82–6.78 (m, 1 H), 3.82 (s, 3 H), 2.40 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 156.7, 144.1, 139.5, 132.0, 129.1, 123.7, 122.9, 122.6, 120.6, 117.7, 117.3, 111.7, 110.9, 55.4, 9.5 ppm. HRMS (ESI): calcd. for $C_{15}H_{15}N_2O$ [M + H]$^+$ 239.1179; found 239.1176.

**2-(4-Methoxyphenyl)-3-methylimidazo[1,2-*a*]pyridine (4aeb):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.87 (d, $J$ = 6.8 Hz, 1 H), 7.72 (d, $J$ = 8.4 Hz, 2 H), 7.62 (dd, $J$ = 9.2, $J$ = 0.8 Hz, 1 H), 7.18–7.14 (m, 1 H), 7.01 (d, $J$ = 8.4 Hz, 2 H), 6.83 (t, $J$ = 6.8 Hz, 1 H), 3.86 (s, 3 H), 2.61 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 159.1, 144.2, 142.2, 129.5, 127.4, 123.4, 122.7, 117.2, 115.2, 114.0, 111.9, 55.3, 9.6 ppm. HRMS (ESI): calcd. for $C_{15}H_{15}N_2O$ [M + H]$^+$ 239.1179; found 239.1185.

**2-(3,4-Dimethoxyphenyl)-3-methylimidazo[1,2-*a*]pyridine (4afb):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.89 (d, $J$ = 6.8 Hz, 1 H), 7.66 (d, $J$ = 9.2 Hz, 1 H), 7.44 (d, $J$ = 2.0 Hz, 1 H), 7.27–7.25 (m, 1 H), 7.20–7.16 (m, 1 H), 6.96 (d, $J$ = 8.0 Hz, 1 H), 6.86 (t, $J$ = 6.8 Hz, 1 H), 3.99 (s, 3 H), 3.93 (s, 3 H), 2.63 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 149.1, 148.6, 144.1, 142.1, 127.5, 123.6, 122.7, 120.6, 117.1, 115.3, 112.1, 111.6, 111.0, 56.0, 55.9, 9.6 ppm. HRMS (ESI): calcd. for $C_{16}H_{17}N_2O_2$ [M + H]$^+$ 269.1285; found 269.1290.

**2-(2-Chlorophenyl)-3-methylimidazo[1,2-*a*]pyridine (4agb):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.92 (d, $J$ = 6.8 Hz, 1 H), 7.67 (d, $J$ = 8.8 Hz, 1 H), 7.58–7.56 (m, 1 H), 7.51–7.48 (m, 1 H), 7.37–7.32 (m, 2 H), 7.23–7.19 (m, 1 H), 6.91–6.87 (m, 1 H), 2.44 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.1, 140.3, 133.64, 133.56, 132.6, 129.7, 129.3, 126.6, 123.7, 122.9, 117.8, 117.6, 112.3, 9.7 ppm. HRMS (ESI): calcd. for $C_{14}H_{12}ClN_2$ [M + H]$^+$ 243.0684; found 243.0681.

**2-(3-Chlorophenyl)-3-methylimidazo[1,2-*a*]pyridine (4ahb):** Yellow solid, m.p. 133–135 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.91 (d, $J$ = 6.8 Hz, 1 H), 7.82 (s, 1 H), 7.69–7.63 (m, 2 H), 7.41–7.38 (m, 1 H), 7.32 (d, $J$ = 8.0 Hz, 1 H), 7.22–7.18 (m, 1 H), 6.89–6.86 (m, 1 H), 2.65 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.4, 141.0, 136.7, 134.5, 129.7, 128.3, 127.4, 126.3, 123.9, 122.9, 117.5, 116.3, 112.3, 9.6 ppm. HRMS (ESI): calcd. for $C_{14}H_{12}ClN_2$ [M + H]$^+$ 243.0684; found 243.0685.

**2-(4-Chlorophenyl)-3-methylimidazo[1,2-*a*]pyridine (4aib):** White solid, m.p. 118–120 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.88 (d, $J$ = 7.2 Hz, 1 H), 7.72 (d, $J$ = 6.8 Hz, 2 H), 7.62 (d, $J$ = 8.8 Hz, 1 H), 7.42 (d, $J$ = 6.8 Hz, 2 H), 7.19–7.16 (m, 1 H), 6.84 (t, $J$ = 6.8 Hz, 1 H), 2.61 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.4, 141.2, 133.3, 133.2, 129.5, 128.6, 123.7, 122.8, 117.4, 116.0, 112.2, 9.6 ppm. HRMS (ESI): calcd. for $C_{14}H_{12}ClN_2$ [M + H]$^+$ 243.0684; found 243.0683.

**2-(2-Bromophenyl)-3-methylimidazo[1,2-*a*]pyridine (4ajb):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.90 (d, $J$ = 6.8 Hz, 1 H), 7.69–7.63 (m, 2 H), 7.51 (dd, $J$ = 7.6, $J$ = 1.6 Hz, 1 H), 7.40–7.36 (m, 1 H), 7.28–7.22 (m, 1 H), 7.21–7.17 (m, 1 H), 6.87 (td, $J$ = 6.8, $J$ = 0.8 Hz, 1 H), 2.42 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 143.9, 142.0, 138.3, 135.7, 132.7, 132.6, 129.4, 127.1, 123.8, 123.5, 122.9, 117.5, 112.1, 9.6 ppm. HRMS (ESI): calcd. for $C_{14}H_{12}BrN_2$ [M + H]$^+$ 287.0178; found 287.0181.

**2-(3-Bromophenyl)-3-methylimidazo[1,2-*a*]pyridine (4akb):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.97 (s, 1 H), 7.91 (d, $J$ = 7.2 Hz, 1 H), 7.72–7.70 (m, 1 H), 7.65–7.62 (m, 1 H), 7.49–7.46 (m, 1 H), 7.34–7.30 (m, 1 H), 7.22–7.18 (m, 1 H), 6.87 (td, $J$ = 6.8, $J$ = 1.2 Hz, 1 H), 2.64 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.3, 140.7, 136.7, 131.1, 130.3, 129.99, 129.95, 126.8,

124.0, 122.9, 122.6, 117.3, 112.3, 9.5 ppm. HRMS (ESI): calcd. for $C_{14}H_{12}BrN_2$ [M + H]$^+$ 287.0178; found 287.0179.

**2-(4-Bromophenyl)-3-methylimidazo[1,2-*a*]pyridine (4alb):** Yellow solid, m.p. 108–110 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.89 (d, $J$ = 7.2 Hz, 1 H), 7.69–7.67 (m, 2 H), 7.63 (d, $J$ = 9.2 Hz, 1 H), 7.60–7.58 (m, 2 H), 7.21–7.17 (m, 1 H), 6.86 (td, $J$ = 6.8, $J$ = 0.8 Hz, 1 H), 2.62 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.4, 141.3, 133.8, 131.6, 129.8, 123.8, 122.9, 121.5, 117.4, 116.0, 112.2, 9.7 ppm. HRMS (ESI): calcd. for $C_{14}H_{12}BrN_2$ [M + H]$^+$ 287.0178; found 287.0175.

**2-(2-Fluorophenyl)-3-methylimidazo[1,2-*a*]pyridine (4amb):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.88–7.85 (m, 1 H), 7.76–7.72 (m, 1 H), 7.65–7.62 (m, 1 H), 7.38–7.31 (m, 1 H), 7.29–7.22 (m, 1 H), 7.18–7.14 (m, 2 H), 6.83–6.79 (m, 1 H), 2.46 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 160.7, 158.3, 144.4, 137.1, 131.82 (d, $J$ = 3.0 Hz), 129.2 (d, $J$ = 9.0 Hz), 124.04 (d, $J$ = 4.0 Hz), 123.3, 122.6, 117.8, 117.2, 115.6 (d, $J$ = 22.0 Hz), 111.8, 9.05 (d, $J$ = 6.0 Hz) ppm. HRMS (ESI): calcd. for $C_{14}H_{12}FN_2$ [M + H]$^+$ 227.0991; found 227.0986.

**3-Methyl-2-(naphthalen-1-yl)imidazo[1,2-*a*]pyridine (4anb):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 8.08 (d, $J$ = 8.0 Hz, 1 H), 7.95 (d, $J$ = 6.4 Hz, 1 H), 7.91–7.89 (m, 2 H), 7.70 (d, $J$ = 9.2 Hz, 1 H), 7.60–7.53 (m, 2 H), 7.51–7.43 (m, 2 H), 7.26–7.20 (m, 1 H), 6.91–6.88 (m, 1 H), 2.43 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.4, 142.1, 133.8, 132.3, 132.2, 128.4, 128.3, 128.1, 126.4, 126.2, 125.7, 125.1, 123.4, 122.9, 117.7, 117.6, 112.0, 9.3 ppm. HRMS (ESI): calcd. for $C_{18}H_{15}N_2$ [M + H]$^+$ 259.1230; found 259.1237.

**2-(Furan-2-yl)-3-methylimidazo[1,2-*a*]pyridine (4aob):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.86 (d, $J$ = 6.8 Hz, 1 H), 7.59 (d, $J$ = 9.2 Hz, 1 H), 7.52 (d, $J$ = 0.8 Hz, 1 H), 7.18–7.14 (m, 1 H), 6.87–6.81 (m, 2 H), 6.52 (q, $J$ = 1.6 Hz, 1 H), 2.70 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 147.3, 144.5, 141.9, 138.5, 123.9, 122.7, 119.6, 117.2, 112.1, 111.3, 107.2, 9.1 ppm. HRMS (ESI): calcd. for $C_{12}H_{11}N_2O$ [M + H]$^+$ 199.0866; found 199.0870.

**2-(2,4-Dichlorophenyl)-3-methylimidazo[1,2-*a*]pyridine (4apb):** Yellow solid, m.p. 128–130 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.90 (d, $J$ = 6.8 Hz, 1 H), 7.63 (d, $J$ = 9.2 Hz, 1 H), 7.51–7.49 (m, 2 H), 7.33 (dd, $J$ = 8.0, $J$ = 2.0 Hz, 1 H), 7.22–7.17 (m, 1 H), 6.88–6.85 (m, 1 H), 2.41 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.1, 139.4, 134.23, 134.15, 133.3, 132.4, 129.3, 126.8, 123.5, 122.8, 117.8, 117.4, 112.1, 9.5 ppm. HRMS (ESI): calcd. for $C_{14}H_{11}Cl_2N_2$ [M + H]$^+$ 277.0294; found 277.0297.

**4-(3-Methylimidazo[1,2-*a*]pyridin-2-yl)benzonitrile (4aqb):** Pale-yellow solid, m.p. 112–114 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.92 (d, $J$ = 8.4 Hz, 3 H), 7.74–7.72 (m, 2 H), 7.64–7.62 (m, 1 H), 7.25–7.20 (m, 1 H), 6.89 (td, $J$ = 6.8, $J$ = 0.8 Hz, 1 H), 2.66 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.6, 140.3, 139.5, 132.2, 128.5, 124.3, 123.0, 119.0, 117.6, 117.2, 112.5, 110.5, 9.7 ppm. HRMS (ESI): calcd. for $C_{15}H_{12}N_3$ [M + H]$^+$ 234.1026; found 234.1020.

**2-(2,6-Dimethylhept-5-en-1-yl)-3-methylimidazo[1,2-*a*]pyridine (4arb):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.79 (d, $J$ = 6.8 Hz, 1 H), 7.54 (d, $J$ = 8.8 Hz, 1 H), 7.11–7.07 (m, 1 H), 6.79–6.76 (m, 1 H), 5.10 (t, $J$ = 6.8 Hz, 1 H), 2.76 (dd, $J$ = 14.0, $J$ = 5.6 Hz, 1 H), 2.53 (dd, $J$ = 13.6, $J$ = 8.4 Hz, 1 H), 2.39 (s, 3 H), 2.09–1.98 (m, 3 H), 1.66 (s, 3 H), 1.59 (s, 3 H), 1.49–1.40 (m, 2 H), 0.90 (d, $J$ = 6.8 Hz, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 143.9, 143.0, 130.9, 124.9, 122.6, 122.4, 116.7, 115.9, 111.3, 37.1, 35.3, 33.8, 25.7, 25.6, 19.4, 17.6, 8.4 ppm. HRMS (ESI): calcd. for $C_{17}H_{25}N_2$ [M + H]$^+$ 257.2012; found 257.2016.

**3,8-Dimethyl-2-phenylimidazo[1,2-*a*]pyridine (4bab):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.80–7.78 (m, 2 H), 7.75 (d, $J$ = 6.8 Hz, 1 H), 7.48–7.44 (m, 2 H), 7.35–7.32 (m, 1 H), 7.96 (d, $J$ = 6.8 Hz, 1 H), 6.75 (t, $J$ = 6.8 Hz, 1 H), 2.66 (s, 3 H), 2.60 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.8, 142.0, 135.1, 128.5, 128.4, 127.3, 127.2, 122.3, 120.7, 116.2, 112.0, 17.1, 9.7 ppm. HRMS (ESI): calcd. for $C_{15}H_{15}N_2$ [M + H]$^+$ 223.1230; found 223.1232.

**3,7-Dimethyl-2-phenylimidazo[1,2-*a*]pyridine (4cab):** White solid, m.p. 173–175 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.78 (dd, $J$ = 8.4 Hz, $J$ = 1.2 Hz, 2 H), 7.74 (d, $J$ = 7.2 Hz, 1 H), 7.47–7.43 (m, 2 H), 7.37 (s, 1 H), 7.35–7.30 (m, 1 H), 6.65–6.63 (m, 1 H), 2.58 (s, 3 H), 2.39 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.7, 141.9, 135.0, 134.3, 128.4, 128.2, 127.1, 122.0, 115.7, 115.2, 114.5, 21.2, 9.5 ppm. HRMS (ESI): calcd. for $C_{15}H_{15}N_2$ [M + H]$^+$ 223.1230; found 223.1226.

**3,6-Dimethyl-2-phenylimidazo[1,2-*a*]pyridine (4dab):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.77 (d, $J$ = 8.0 Hz, 2 H), 7.61 (s, 1 H), 7.51 (d, $J$ = 9.2 Hz, 1 H), 7.44 (t, $J$ = 7.6 Hz, 2 H), 7.34–7.30 (m, 1 H), 6.98 (d, $J$ = 9.2 Hz, 1 H), 2.55 (s, 3 H), 2.32 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 143.3, 142.1, 134.9, 128.3, 128.1, 127.1, 126.5, 121.4, 120.5, 116.6, 115.5, 18.3, 9.5 ppm. HRMS (ESI): calcd. for $C_{15}H_{15}N_2$ [M + H]$^+$ 223.1230; found 223.1228.

**3,5-Dimethyl-2-phenylimidazo[1,2-*a*]pyridine (4eab):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.69–7.66 (m, 2 H), 7.47–7.43 (m, 3 H), 7.37–7.33 (m, 1 H), 6.99 (dd, $J$ = 8.8, $J$ = 6.8 Hz, 1 H), 6.44 (d, $J$ = 6.8 Hz, 1 H), 2.91 (s, 6 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 146.1, 143.9, 136.2, 134.9, 129.1, 128.3, 127.4, 124.1, 117.8, 115.9, 113.3, 20.8, 13.5 ppm. HRMS (ESI): calcd. for $C_{15}H_{15}N_2$ [M + H]$^+$ 223.1230; found 223.1233.

**7-Chloro-3-methyl-2-phenylimidazo[1,2-*a*]pyridine (4fab):** Pale-yellow solid, m.p. 124–126 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.75 (d, $J$ = 8.0 Hz, 3 H), 7.60 (s, 1 H), 7.45 (t, $J$ = 7.6 Hz, 2 H), 7.36–7.32 (m, 1 H), 6.77 (dd, $J$ = 7.2, $J$ = 2.0 Hz, 1 H), 2.56 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 143.8, 143.1, 134.3, 129.9, 128.5, 128.2, 127.5, 123.0, 116.1, 116.0, 113.4, 9.5 ppm. HRMS (ESI): calcd. for $C_{14}H_{12}ClN_2$ [M + H]$^+$ 243.0684; found 243.0688.

**6-Chloro-3-methyl-2-phenylimidazo[1,2-*a*]pyridine (4gab):** Yellow solid, m.p. 125–127 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.91 (s, 1 H), 7.78–7.76 (m, 2 H), 7.57 (dd, $J$ = 9.6, $J$ = 0.4 Hz, 1 H), 7.48–7.44 (m, 2 H), 7.38–7.33 (m, 1 H), 7.14–7.11 (m, 1 H), 2.60 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 143.4, 142.5, 134.2, 128.5, 128.2, 127.6, 124.8, 120.7, 120.3, 117.7, 116.5, 9.6 ppm. HRMS (ESI): calcd. for $C_{14}H_{12}ClN_2$ [M + H]$^+$ 243.0684; found 243.0678.

**6-Bromo-3-methyl-2-phenylimidazo[1,2-*a*]pyridine (4hab):** Yellow solid, m.p. 73–75 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 8.00 (s, 1 H), 7.76 (d, $J$ = 7.6 Hz, 2 H), 7.51 (d, $J$ = 9.6 Hz, 1 H), 7.46 (t, $J$ = 7.6 Hz, 2 H), 7.37–7.33 (m, 1 H), 7.20 (dd, $J$ = 9.2, $J$ = 1.2 Hz, 1 H), 2.59 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 143.3, 142.7, 134.3, 128.5, 128.2, 127.6, 126.7, 123.0, 118.0, 116.3, 106.7, 9.6 ppm. HRMS (ESI): calcd. for $C_{14}H_{12}BrN_2$ [M + H]$^+$ 287.0178; found 287.0173.

**6-Fluoro-3-methyl-2-phenylimidazo[1,2-*a*]pyridine (4iab):** Yellow solid, m.p. 88–90 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.75 (d, $J$ = 9.6 Hz, 3 H), 7.59–7.55 (m, 1 H), 7.46–7.42 (m, 2 H), 7.35–7.31 (m, 1 H), 7.07–7.02 (m, 1 H), 2.55 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 154.4, 152.0, 143.8, 141.9, 134.4, 128.3 (d, $J$ = 32.0 Hz), 127.4, 117.7 (d, $J$ = 9.0 Hz), 117.3, 115.2 (d, $J$ = 25.0 Hz),

109.4 (d, $J$ = 41.0 Hz), 9.6 ppm. HRMS (ESI): calcd. for $C_{14}H_{12}FN_2$ $[M + H]^+$ 227.0991; found 227.0997.

**Ethyl-3-methyl-2-phenylimidazo[1,2-*a*]pyridine-7-carboxylate (4jab):** Yellow solid, m.p. 130–133 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 8.37 (s, 1 H), 7.86 (d, $J$ = 6.8 Hz, 1 H), 7.79 (d, $J$ = 7.2 Hz, 2 H), 7.48–7.45 (m, 2 H), 7.42–7.34 (m, 2 H), 4.40 (q, $J$ = 7.2 Hz, 2 H), 2.62 (s, 3 H), 1.41 (t, $J$ = 7.2 Hz, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 165.3, 145.1, 143.1, 134.1, 128.6, 128.2, 124.9, 122.0, 119.9, 119.6, 117.9, 111.4, 61.3, 13.9, 9.6 ppm. HRMS (ESI): calcd. for $C_{17}H_{17}N_2O_2$ $[M + H]^+$ 281.1285; found 281.1287.

**8-(Benzyloxy)-3-methyl-2-phenylimidazo[1,2-*a*]pyridine (4kab):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.83 (d, $J$ = 7.2 Hz, 2 H), 7.50–7.48 (m, 3 H), 7.44 (t, $J$ = 7.6 Hz, 2 H), 7.38–7.32 (m, 3 H), 7.30–7.25 (m, 1 H), 6.62 (t, $J$ = 7.2 Hz, 1 H), 6.42 (d, $J$ = 7.6 Hz, 1 H), 5.39 (s, 2 H), 2.57 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 147.8, 141.6, 138.9, 136.5, 134.8, 128.52, 128.48, 128.3, 127.9, 127.8, 127.1, 116.8, 116.0, 111.7, 102.4, 70.6, 9.8 ppm. HRMS (ESI): calcd. for $C_{21}H_{19}N_2O$ $[M + H]^+$ 315.1492; found 315.1494.

**6-Bromo-3,5-dimethyl-2-phenylimidazo[1,2-*a*]pyridine (4lab):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.66–7.64 (m, 2 H), 7.47–7.44 (m, 2 H), 7.38–7.31 (m, 2 H), 7.21 (d, $J$ = 9.2 Hz, 1 H), 3.05 (s, 3 H), 2.89 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 145.1, 144.9, 134.5, 134.3, 129.2, 128.9, 128.2, 127.9, 127.8, 119.2, 109.3, 19.6, 14.5 ppm. HRMS (ESI): calcd. for $C_{15}H_{14}BrN_2$ $[M + H]^+$ 301.0334; found 301.0340.

**Typical Procedure for the Preparation of 2-Arylimidazo[1,2-*a*]pyridine Derivatives:** A test tube was charged with **1** (0.5 mmol), **2** (0.6 mmol), TBAI (0.5 mmol) and $FeCl_3$ (8.13 mg, 0.05 mmol). Then **3a** (10 equiv. of $MeNO_2$) was added to the reaction system. The reaction was stirred at 90 °C under air for 4 h. After cooling to room temperature, the solvent diluted with ethyl acetate (10 mL) was evaporated in vacuo, and the residues were purified by column chromatography (petroleum ether/EtOAc) to afford pure **4aaa**.

**2-Phenylimidazo[1,2-*a*]pyridine (4aaa):** White solid, m.p. 106–108 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 8.07 (d, $J$ = 6.8 Hz, 1 H), 7.95 (d, $J$ = 7.2 Hz, 2 H), 7.82 (s, 1 H), 7.62 (d, $J$ = 9.2 Hz, 1 H), 7.43 (t, $J$ = 7.6 Hz, 2 H), 7.34–7.30 (m, 1 H), 7.16–7.12 (m, 1 H), 6.74 (t, $J$ = 6.8 Hz, 1 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 145.7, 145.6, 133.7, 128.7, 127.9, 126.0, 125.5, 124.6, 117.4, 112.3, 108.1 ppm. HRMS (ESI): calcd. for $C_{13}H_{11}N_2$ $[M + H]^+$ 195.0917; found 195.0921.

**2-(*p*-Tolyl)imidazo[1,2-*a*]pyridine (4aba):** Yellow solid, m.p. 114–116 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 8.08 (d, $J$ = 6.8 Hz, 1 H), 7.85 (d, $J$ = 8.0 Hz, 2 H), 7.81 (s, 1 H), 7.62 (d, $J$ = 9.2 Hz, 1 H), 7.25 (d, $J$ = 8.0 Hz, 2 H), 7.17–7.13 (m, 1 H), 6.75 (t, $J$ = 6.8 Hz, 1 H), 2.39 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 145.9, 145.6, 137.8, 130.9, 129.4, 125.9, 125.5, 124.5, 117.4, 112.3, 107.7, 21.3 ppm. HRMS (ESI): calcd. for $C_{14}H_{13}N_2$ $[M + H]^+$ 209.1073; found 209.1079.

**2-(2-Methoxyphenyl)imidazo[1,2-*a*]pyridine (4ada):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 8.41 (dd, $J$ = 7.6, $J$ = 1.6 Hz, 1 H), 8.16 (s, 1 H), 8.06 (d, $J$ = 6.8 Hz, 1 H), 7.61–7.59 (m, 1 H), 7.32–7.27 (m, 1 H), 7.13–7.08 (m, 2 H), 6.97 (d, $J$ = 8.4 Hz, 1 H), 6.69 (t, $J$ = 6.8 Hz, 1 H), 3.95 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 156.6, 144.2, 140.9, 128.6, 128.5, 125.5, 124.4, 122.1, 120.8, 117.0, 112.4, 111.8, 110.7, 55.2 ppm. HRMS (ESI): calcd. for $C_{14}H_{13}N_2O$ $[M + H]^+$ 225.1023; found 225.1026.

**2-(3-Bromophenyl)imidazo[1,2-*a*]pyridine (4aka):** Yellow solid, m.p. 104–106 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 8.13–8.12 (m, 2 H), 7.88 (s, 1 H), 7.86 (s, 1 H), 7.63 (d, $J$ = 9.2 Hz, 1 H), 7.45 (dd, $J$ = 8.0, $J$ = 0.8 Hz, 1 H), 7.30 (d, $J$ = 8.0 Hz, 1 H), 7.22–7.18 (m, 1 H), 6.81 (td, $J$ = 6.8, $J$ = 0.8 Hz, 1 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 145.7, 144.2, 135.8, 130.8, 130.2, 128.9, 125.7, 125.1, 124.5, 122.9, 117.6, 112.7, 108.5 ppm. HRMS (ESI): calcd. for $C_{13}H_{10}BrN_2$ $[M + H]^+$ 273.0021; found 273.0018.

**2-(2-Fluorophenyl)imidazo[1,2-*a*]pyridine (4ama):** Yellow solid, m.p. 80–82 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 8.31 (td, $J$ = 7.6, $J$ = 1.6 Hz, 1 H), 8.12 (d, $J$ = 6.8 Hz, 1 H), 8.04 (d, $J$ = 4.0 Hz, 1 H), 7.62 (d, $J$ = 9.2 Hz, 1 H), 7.30–7.22 (m, 2 H), 7.20–7.12 (m, 2 H), 6.78 (t, $J$ = 6.8 Hz, 1 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 161.5, 159.0, 144.8, 139.0, 128.9 (d, $J$ = 9.0 Hz), 128.7 (d, $J$ = 3.0 Hz), 125.7, 125.0, 124.5 (d, $J$ = 4.0 Hz), 117.3, 115.7, 115.5, 112.5 ppm. HRMS (ESI): calcd. for $C_{13}H_{10}FN_2$ $[M + H]^+$ 213.0835; found 213.0837.

**4-(Imidazo[1,2-*a*]pyridin-2-yl)benzonitrile (4aqa):** Yellow solid, m.p. 146–148 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 8.13 (d, $J$ = 6.8 Hz, 1 H), 8.02 (d, $J$ = 8.0 Hz, 2 H), 7.93 (s, 1 H), 7.68 (d, $J$ = 8.0 Hz, 2 H), 7.60 (d, $J$ = 9.2 Hz, 1 H), 7.22 (d, $J$ = 8.0 Hz, 1 H), 6.83 (d, $J$ = 6.8 Hz, 1 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 145.8, 143.4, 138.1, 132.4, 126.2, 125.8, 125.5, 119.0, 117.6, 113.0, 110.9, 109.5 ppm. HRMS (ESI): calcd. for $C_{14}H_{10}N_3$ $[M + H]^+$ 220.0869; found 220.0874.

**2-[4-(Methylthio)phenyl]imidazo[1,2-*a*]pyridine (4asa):** Yellow solid, m.p. 138–140 °C. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 8.09 (d, $J$ = 6.8 Hz, 1 H), 7.87 (d, $J$ = 8.8 Hz, 2 H), 7.82 (s, 1 H), 7.62 (d, $J$ = 9.6 Hz, 1 H), 7.31 (d, $J$ = 8.4 Hz, 2 H), 7.18–7.14 (m, 1 H), 6.76 (td, $J$ = 6.8, $J$ = 0.8 Hz, 1 H), 2.52 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 145.7, 145.4, 138.2, 130.7, 126.8, 126.4, 125.5, 124.6, 117.4, 112.4, 107.8, 15.8 ppm. HRMS (ESI): calcd. for $C_{14}H_{13}N_2S$ $[M + H]^+$ 241.0794; found 241.0798.

**8-Methyl-2-phenylimidazo[1,2-*a*]pyridine (4baa):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.97–7.94 (m, 3 H), 7.81 (s, 1 H), 7.43 (t, $J$ = 7.6 Hz, 2 H), 7.31 (t, $J$ = 7.6 Hz, 1 H), 6.93 (d, $J$ = 6.8 Hz, 1 H), 6.65 (t, $J$ = 6.8 Hz, 1 H), 2.66 (s, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 146.1, 145.1, 134.0, 128.6, 127.7, 127.5, 126.1, 123.4, 123.2, 112.3, 108.5, 17.1 ppm. HRMS (ESI): calcd. for $C_{14}H_{13}N_2$ $[M + H]^+$ 209.1073; found 209.1068.

**Typical Procedure for the Preparation of 3-Ethyl-2-arylimidazo[1,2-*a*]-pyridine Derivatives:** A test tube was charged with **1a** (0.5 mmol), **2** (0.6 mmol) and $FeCl_3$ (8.13 mg, 0.05 mmol). Then **3c** (10 equiv. of 1-nitropropane) was added to the reaction system. The reaction was stirred at 90 °C under air for 4 h. After cooling to room temperature, the solvent diluted with ethyl acetate (10 mL) was evaporated in vacuo, and the residues were purified by column chromatography (petroleum ether/EtOAc) to afford pure **4aac**.

**3-Ethyl-2-phenylimidazo[1,2-*a*]pyridine (4aac):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.93 (d, $J$ = 6.8 Hz, 1 H), 7.80–7.78 (m, 2 H), 7.64 (d, $J$ = 9.2 Hz, 1 H), 7.48–7.44 (m, 2 H), 7.36–7.33 (m, 1 H), 7.16–7.12 (m, 1 H), 6.79 (t, $J$ = 6.8 Hz, 1 H), 3.09 (q, $J$ = 7.6 Hz, 2 H), 1.34 (t, $J$ = 7.6 Hz, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.1, 141.4, 134.5, 128.4, 128.1, 127.4, 123.7, 122.8, 121.7, 117.4, 112.1, 16.9, 12.1 ppm. HRMS (ESI): calcd. for $C_{15}H_{15}N_2$ $[M + H]^+$ 223.1230; found 223.1238.

**3-Ethyl-2-(*p*-tolyl)imidazo[1,2-*a*]pyridine (4abc):** Yellow viscous oil. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 7.94 (d, $J$ = 6.8 Hz, 1 H), 7.68 (d, $J$ = 8.4 Hz, 2 H), 7.64 (d, $J$ = 9.2 Hz, 1 H), 7.29–7.27 (m, 2 H), 7.18–7.12 (m, 1 H), 6.81 (td, $J$ = 6.8, $J$ = 0.8 Hz, 1 H), 3.10 (q, $J$ = 7.6 Hz, 2 H), 2.41 (s, 3 H), 1.35 (t, $J$ = 7.6 Hz, 3 H) ppm. $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 144.2, 141.8, 137.1, 131.8, 129.2, 128.0, 123.5, 122.8, 121.4, 117.5, 111.9, 21.2, 17.0, 12.2 ppm.

HRMS (ESI): calcd. for $C_{16}H_{17}N_2$ $[M + H]^+$ 237.1386; found 237.1381.

**3-Ethyl-2-(4-methoxyphenyl)imidazo[1,2-*a*]pyridine (4aec):** Yellow viscous oil. $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ = 7.92 (d, $J$ = 6.8 Hz, 1 H), 7.72 (d, $J$ = 8.8 Hz, 2 H), 7.62 (d, $J$ = 9.2 Hz, 1 H), 7.16–7.12 (m, 1 H), 7.00 (d, $J$ = 8.8 Hz, 2 H), 6.80 (t, $J$ = 6.8 Hz, 1 H), 3.85 (s, 3 H), 3.07 (q, $J$ = 7.6 Hz, 2 H), 1.34 (t, $J$ = 7.6 Hz, 3 H) ppm. $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ = 159.0, 144.1, 141.5, 129.2, 127.3, 123.3, 122.7, 121.0, 117.3, 113.9, 111.8, 55.2, 17.0, 12.1 ppm. HRMS (ESI): calcd. for $C_{16}H_{17}N_2O$ $[M + H]^+$ 253.1336; found 253.1334.

**2-(3,4-Dimethoxyphenyl)-3-ethylimidazo[1,2-*a*]pyridine (4afc):** Yellow viscous oil. $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ = 7.95 (d, $J$ = 6.8 Hz, 1 H), 7.66 (d, $J$ = 9.2 Hz, 1 H), 7.42 (d, $J$ = 1.6 Hz, 1 H), 7.26–7.23 (m, 1 H), 7.19–7.15 (m, 1 H), 6.96 (d, $J$ = 8.0 Hz, 1 H), 6.84 (t, $J$ = 6.8 Hz, 1 H), 3.97 (s, 3 H), 3.94 (s, 3 H), 3.10 (q, $J$ = 7.6 Hz, 2 H), 1.36 (t, $J$ = 7.6 Hz, 3 H) ppm. $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ = 149.0, 148.5, 144.1, 141.5, 127.5, 123.6, 122.7, 121.2, 120.2, 117.3, 112.0, 111.4, 110.9, 55.82, 55.80, 17.0, 12.1 ppm. HRMS (ESI): calcd. for $C_{17}H_{19}N_2O_2$ $[M + H]^+$ 283.1441; found 283.1448.

**2-(4-Chlorophenyl)-3-ethylimidazo[1,2-*a*]pyridine (4aic):** Yellow solid, m.p. 100–102 °C. $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ = 7.93 (d, $J$ = 6.8 Hz, 1 H), 7.71 (d, $J$ = 8.4 Hz, 2 H), 7.62 (d, $J$ = 9.2 Hz, 1 H), 7.42 (d, $J$ = 8.8 Hz, 2 H), 7.19–7.15 (m, 1 H), 6.82 (t, $J$ = 6.8 Hz, 1 H), 3.06 (q, $J$ = 7.6 Hz, 2 H), 1.33 (t, $J$ = 7.6 Hz, 3 H) ppm. $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ = 144.3, 140.5, 133.3, 133.2, 129.2, 128.6, 123.8, 122.8, 121.8, 117.5, 112.1, 16.9, 12.1 ppm. HRMS (ESI): calcd. for $C_{15}H_{14}ClN_2$ $[M + H]^+$ 257.0840; found 257.0845.

**2-(2-Bromophenyl)-3-ethylimidazo[1,2-*a*]pyridine (4ajc):** Yellow solid, m.p. 76–78 °C. $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ = 7.96 (d, $J$ = 6.8 Hz, 1 H), 7.68–7.62 (m, 2 H), 7.45 (dd, $J$ = 7.6, $J$ = 1.2 Hz, 1 H), 7.36 (td, $J$ = 7.6, $J$ = 1.2 Hz, 1 H), 7.25–7.21 (m, 1 H), 7.17–7.13 (m, 1 H), 6.81 (td, $J$ = 6.8, $J$ = 1.2 Hz, 1 H), 2.87 (q, $J$ = 7.6 Hz, 2 H), 1.17 (t, $J$ = 7.6 Hz, 3 H) ppm. $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ = 143.8, 141.5, 136.0, 132.5, 132.2, 129.3, 126.8, 124.0, 123.3, 123.0, 122.7, 117.7, 111.9, 16.8, 11.6 ppm. HRMS (ESI): calcd. for $C_{15}H_{14}BrN_2$ $[M + H]^+$ 301.0334; found 301.0337.

**3-Ethyl-2-(2-fluorophenyl)imidazo[1,2-*a*]pyridine (4amc):** Yellow viscous oil. $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ = 7.95 (d, $J$ = 7.2 Hz, 1 H), 7.71–7.63 (m, 2 H), 7.37–7.32 (m, 1 H), 7.26–7.22 (m, 1 H), 7.18–7.13 (m, 2 H), 6.82–6.79 (m, 1 H), 2.94 (q, $J$ = 7.6 Hz, 2 H), 1.26 (t, $J$ = 7.6 Hz, 3 H) ppm. $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ = 160.9, 158.5, 144.5, 136.4, 132.0 (d, $J$ = 3.0 Hz), 129.3 (d, $J$ = 8.0 Hz), 124.1 (d, $J$ = 3.0 Hz), 123.4 (d, $J$ = 5.0 Hz), 123.0, 117.6, 115.7, 115.5, 111.9, 16.9, 11.5 ppm. HRMS (ESI): calcd. for $C_{15}H_{14}FN_2$ $[M + H]^+$ 241.1148; found 241.1144.

**4-(3-Ethylimidazo[1,2-*a*]pyridin-2-yl)benzonitrile (4aqc):** Yellow solid, m.p. 158–160 °C. $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ = 7.98 (d, $J$ = 6.8 Hz, 1 H), 7.91 (d, $J$ = 8.0 Hz, 2 H), 7.74 (d, $J$ = 8.4 Hz, 2 H), 7.63 (d, $J$ = 9.2 Hz, 1 H), 7.24–7.21 (m, 1 H), 6.89 (t, $J$ = 6.8 Hz, 1 H), 3.12 (q, $J$ = 7.6 Hz, 2 H), 1.38 (t, $J$ = 7.6 Hz, 3 H) ppm. $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ = 144.5, 139.6, 139.5, 132.2, 128.3, 124.4, 123.1, 122.9, 119.0, 117.8, 112.5, 110.5, 17.0, 12.1 ppm. HRMS (ESI): calcd. for $C_{16}H_{14}N_3$ $[M + H]^+$ 248.1182; found 248.1185.

**Supporting Information** (see footnote on the first page of this article): $^1H$ and $^{13}C$ NMR spectra of the products.

## Acknowledgments

The authors are grateful for financial support by the Fundamental Research Funds for the Central Universities, P. R. China.

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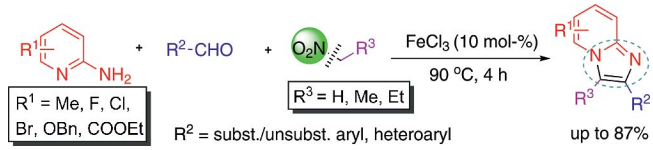
Received: November 5, 2013
Published Online: ■

EurJOC European Journal of Organic Chemistry

**Nitrogen Heterocycles**




An iron(III)-catalyzed one-pot three-component cross-coupling nitration reaction of 2-aminopyridines, aldehydes, and nitroalkane, leading to the straightforward formation of imidazo[1,2-*a*]pyridine derivatives has been reported. In this procedure, the starting materials are commercially available. The system shows good functional-group tolerance and proceeds smoothly in moderate to good yields.



**Keywords:** Synthetic methods / Multicomponent reactions / Nitrogen heterocycles / Iron