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Transition Metal-Mediated C=O and C=C Bond-Forming Reactions: A Regioselective Strategy for the Synthesis of Imidazo[1,2-*a*]pyridines and Imidazo[1,2-*a*]pyrazines

Hua Cao,*[,†] Xiaohang Liu,[‡] Jinqiang Liao,[†] Jianping Huang,[†] Huifang Qiu,[†] Qinlin Chen,[†] and Yaoyi Chen[†]

[†]School of Chemistry and Chemical Engineering, Guangdong Pharmaceutical University, Guangzhou 510006, P.R. China

[‡]BASF Catalyst, 23800 Mercantile Road, Beachwood, Ohio 44122, United States

Supporting Information

ABSTRACT: A novel and convenient transformation for the regiospecific synthesis of functionalized imidazo[1,2-*a*]pyridine aldehydes/ketones and 3-vinyl imidazo[1,2-*a*]pyridines has been developed via copper(I)- and palladium(II)-catalyzed cyclization. The one-pot reaction proceeds smoothly with commercially available catalysts and affords the products in moderate to good yields. It represents an efficient approach for the formation of C−N, C=O, and C=C bonds under mild conditions.

Since the first discovery of stable transition metal carbene complexes by Fischer and Maasböl,[1] metal carbenes have attracted the attention of organic chemists.[2] Significant development of metal carbenes, as highly useful intermediates in organic synthesis, has already taken place, and this has become an important branch and field of organometallic chemistry. Over the past few years, a number of novel transformations have been reported using these versatile metal complexes.[3] Barluenga,[4] Vicente and López,[5] and Wang[6] reported synthesis of furans via carbene complexes. We have also reported a convenient approach to synthesize furans via copper[7] and palladium carbene complexes.[8] In fact, designing new strategies for the selective synthesis of five- or six-membered heterocyclic compounds via metal carbene complexes continues to attract our attention. Currently, our interest is focused on developing a facile transformation to prepare imidazo[1,2-*a*]pyridines via metal carbene complexes.

Imidazo[1,2-*a*]pyridine is a fundamental class of heterocycles and is worthy of our attention for many reasons.[9] Many drugs contain the core structure of imidazo[1,2-*a*]pyridine, such as zolpidem, alpidem, zolimidine, olprinone, saripidem, and necopidem.[10] Therefore, organic chemists have been making extensive efforts to prepare imidazo[1,2-*a*]pyridine derivatives[11] by developing novel and convenient organic transformations.

Recently, a number of successful synthetic strategies have been reported for the construction of imidazo[1,2-*a*]-pyridines.[12] Although extensive investigation in this field has been conducted, the development of new routes is still highly desirable for the synthesis of functionalized imidazo[1,2-*a*]pyridines from readily accessible starting materials in a single operation under mild conditions.[13] Herein, we report the first examples of a one-pot process for the formation of C=O and C=C bonds to prepare functionalized imidazo[1,2-*a*]pyridines via metal carbene complexes.

In our initial studies, 3-phenylpropiolaldehyde (**1a**) and pyridin-2-amine (**2a**) were chosen as the substrates to investigate the formation of imidazo[1,2-*a*]pyridin-3-yl-(phenyl)methanone (**3a**). On the basis of our previous work, a variety of copper catalysts in conjunction with different ligands, solvents, and temperatures were screened, and the results are summarized in Table 1. The desired product **3a** was formed in 12% yield when the reaction carried out using CuCl as a catalyst in CH_2Cl_2 at rt for 12 h (Table 1, entry 1). Inspired by this promising result, various copper salts, such as CuBr, CuI, and Cu_2O, were next screened to study their catalytic efficiency for the synthesis of **3a** (entries 2−4). The results indicated that the copper salts screened did promote the reaction at room temperature. Among them, CuI is a more efficient catalyst than the other tested. Having gained some crucial insight into the effect of different catalysts, further optimization was performed to explore the effect of ligands (entries 5−11). Gratifyingly, Bipy proved to be the best ligand, and the corresponding product **3a** was obtained in good yields, whereas other ligands, such as Py, Phen, DMEDA, TMEDA, DABCO, and PPh_3, disfavored the reaction to varying degrees. Subsequently, the reaction conditions were further optimized by examining various solvents (entries 12−16). A variety of polar and nonpolar solvents were tested, and the results showed that CH_2Cl_2 the best solvent. Further investigation found that the transformation was sensitive to temperature variation. The desired product **3a** was obtained only in 23 and 11% yields when the reactions were carried out at 50 and 80 °C, respectively (entries 17−19). Actually, only trace product was detected when the temperature was increased to 100 °C. Control experiments in the absence of CuI were ineffective, and no product was formed (entries 20−22). No products were

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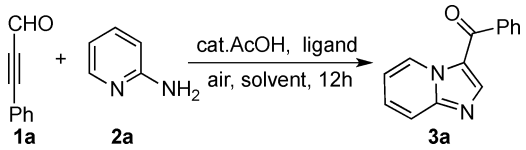
Table 1. Screen of the Reaction Conditions[a]



entry	catalyst	ligand	solvent	T (°C)	yield (%)[b]
1	CuCl		CH_2Cl_2	rt	12
2	CuBr		CH_2Cl_2	rt	19
3	CuI		CH_2Cl_2	rt	35
4	Cu_2O		CH_2Cl_2	rt	26
5	CuI	Py	CH_2Cl_2	rt	46
6	CuI	Bipy	CH_2Cl_2	rt	85 (80)[c]
7	CuI	Phen	CH_2Cl_2	rt	41
8	CuI	DMEDA	CH_2Cl_2	rt	36
9	CuI	TMEDA	CH_2Cl_2	rt	42
10	CuI	DABCO	CH_2Cl_2	rt	30
11	CuI	PPh_3	CH_2Cl_2	rt	27
12	CuI	Bipy	CH_3CN	rt	45
13	CuI	Bipy	toluene	rt	66
14	CuI	Bipy	DMF	rt	73
15	CuI	Bipy	DMSO	rt	71
16	CuI	Bipy	dioxane	rt	80
17	CuI	Bipy	CH_2Cl_2	50	23
18	CuI	Bipy	CH_2Cl_2	80	11
19	CuI	Bipy	CH_2Cl_2	100	trace
20		Bipy	CH_2Cl_2	rt	N.P.
21			CH_2Cl_2	50	N.P.
22			CH_2Cl_2	50	N.P.
23[d]	CuI	Bipy	CH_2Cl_2	rt	

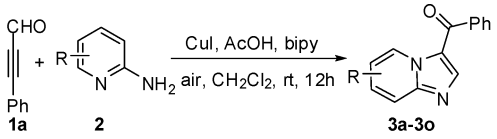
[a]Reaction conditions: **1a** (0.5 mmol), **2a** (0.6 mmol), AcOH (5 mol %), catalyst (5 mol %), ligand (10 mol %), solvents 3 mL; rt to 100 °C for 12 h. [b]GC yield. [c]Isolated yield. [d]Under anaerobic conditions.

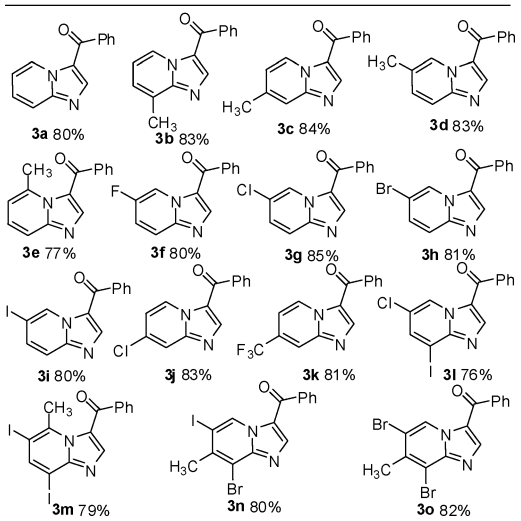
obtained when the reaction was carried out by strictly omitting oxygen in the presence of CuI and bipy in CH_2Cl_2 (entry 23).

Using the mild conditions [CuI (5 mol %), Bipy (10 mol %), 5 mol % AcOH, CH_3CN, rt] established above, the scope of this one-pot copper-catalyzed process was investigated for the synthesis of imidazo[1,2-*a*]pyridines. **1a** was fixed as the substrate to test various substituted pyridin-2-amines, and the results are outlined in Table 2. All of the reactions proceeded smoothly under the optimized conditions and provided the substituted imidazo[1,2-*a*]pyridine derivatives in good yields. It was found that electron-rich (CH_3) and electron-poor (F, CF_3) substituted pyridin-2-amines proved to be suitable for this Cu-catalyzed one-pot process, and the corresponding products were obtained in 77−81% yields. To our delight, the results indicated that F, Cl, Br, and I substituted on the pyridine ring were also well-tolerated under these conditions and led to a beneficial effect on the reaction outcome. It is worth pointing out that multi-halogen-substituted pyridin-2-amines could also be achieved upon carrying out the experiment under the optimized conditions.

After achieving the results given in Table 2, we next investigated this reaction between other substituted propiolaldehydes and various substituted pyridin-2-amines, and the results are summarized in Table 3. First, oct-2-ynal (**1b**) was examined. It was found that the reaction of **1b** with pyridin-2-amines or its derivatives with electron-rich methyl groups at the 3-, 4-, 5- and 6-position were smooth and afforded the corresponding products **4a**−**4e** in 70−79% yields. The halo

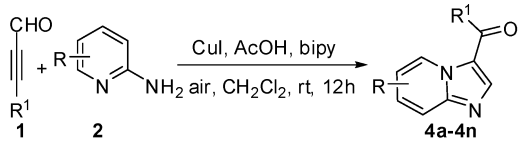
Table 2. Synthesis of Imidazo[1,2-*a*]pyridines from 1a and 2[a]

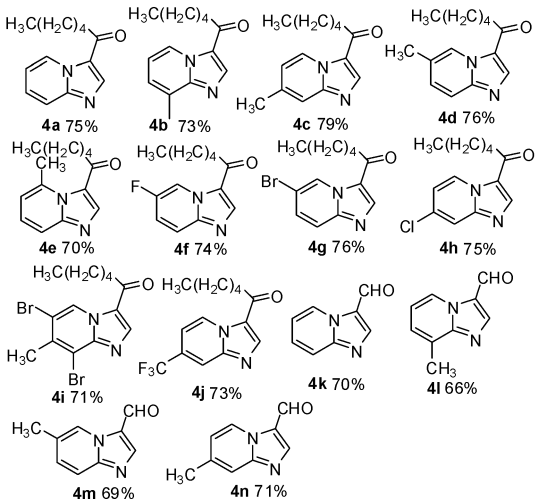




[a]Isolated yields.

Table 3. Formation of Imidazo[1,2-*a*]pyridines[a]





[a]Isolated yields.

groups (chloro, bromo, iodo) remained unaffected under these reaction conditions, and multiple halo groups on the pyridine ring were also well-tolerated. Moreover, substrate **2** with a strong electron-withdrawing CF_3 group did not affect the reactivity and afforded the product in moderate yield. Subsequently, propiolaldehyde **1c** was employed. To our delight, the desired products **4k**−**4p** were formed in good yields under the optimized conditions. All of these results

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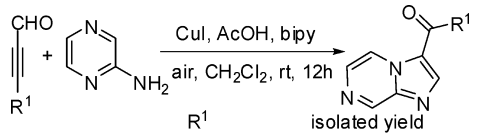
indicated that the CuI-catlayzed transformation is applicable to both aryl and alkyl substituted as well as terminal propiolaldehydes.

For further investigation, pyrazin-2-amine was also tested, and the results are shown in Scheme 1. The reaction of pyrazin-2-amine with **1a** or **1b** performed well and formed the desired products 3-carbonyl imidazo[1,2-*a*]pyrazine (**4o** and **4p**) in moderate yields under the optimized conditions. This represents a novel approach for the construction of a new carbon−oxygen bond to synthesize 3-carbonyl imidazo[1,2-*a*]pyrazine.

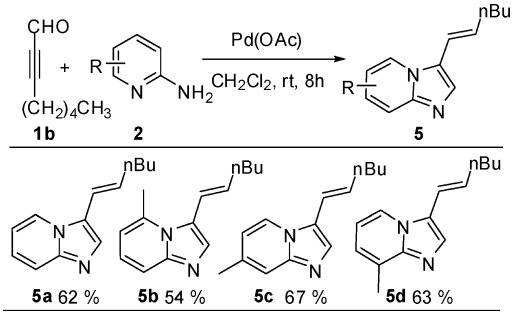
Scheme 1. Synthesis of Imidazo[1,2-*a*]pyrazine



	R^1	isolated yield
4o	Ph	52%
4p	$CH_3(CH_2)_4$	45%

Very recently, we reported the Pd-catalyzed regioselective synthesis of 2-vinyl furans via a palladium carbene complex.[8,14] On the basis of the previous results, we next investigated Pd-catalyzed transformation to form carbon−carbon double bonds for the construction of 3-vinyl imidazo[1,2-*a*]pyridines via a palladium carbene complex (Scheme 2). To our delight, the 3-vinyl imidazo[1,2-*a*]pyridine products were obtained in moderate yields when the reaction was carried out with $Pd(OAc)_2$ in CH_3CN at room temperature for 3 h. Our findings demonstrate a new Pd(OAc)-catalyzed synthetic route to prepare 3-vinyl imidazo[1,2-*a*]pyridines, of which further studies and applications of the transformation are ongoing in our laboratory. (For the proposed mechanism, see the Supporting Information.)

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



[a]Isolated yields

In summary, we report two novel one-pot processes for the preparation of C−N, C═O, and C═C bonds, leading to polysubstituted imidazo[1,2-*a*]pyridines starting from simple and readily available inputs. One process is the CuI-catalyzed synthesis of functionalized imidazo[1,2-*a*]pyridinealdehydes/ketones via copper carbene oxidation using air as the sole oxidant. The other is $Pd(OAc)_2$-catalyzed construction of 3-vinylimidazo[1,2-*a*]pyridines via a 1,2-H shift of palladium carbene complexes. Most of the common functionalities, such as F, Cl, Br, I, and CF_3, are well-tolerated. This one-pot reaction provides an efficient method for the regiospecific synthesis of functionalized imidazo[1,2-*a*]pyridines, which are broadly applicable for the synthesis of biologically active molecules.

EXPERIMENTAL SECTION

^1H and ^{13}C NMR spectra were recorded at 400 and 100 MHz in $CDCl_3$. Mass spectra were recorded by electron ionization. Elemental analyses were performed with a Vario EL element analyzer. GC-MS was obtained using electron ionization. TLC was performed using commercially prepared 100−400 mesh silica gel plates, and visualization was performed at 254 nm.

Synthesis of 3a . 3-Phenylpropiolaldehyde (**1a**, 0.5 mmol), pyridin-2-amine(**2a**, 0.6 mmol), AcOH (5 mol %), CuI (5 mol %), and Bipy (10 mol %) were stirred for 12 h in CH_2Cl_2 (3 mL) at room temperature. After reaction completion, as monitored by TLC and GC-MS analysis, the solvent was then removed, and the crude product was separated by column chromatography (eluted with petroleum ether/ethyl acetate = 2:1) to give a pure sample of **3a** (80%, 88.8 mg).

Synthesis of 5a. Oct-2-ynal (**2a**, 0.5 mmol), pyridin-2-amine (**2a**, 0.6 mmol), AcOH (5 mol %), and $Pd(OAc)_2$ (5 mol %) were stirred for 6 h in CH_2Cl_2 (3 mL) at room temperature. After reaction completion, as monitored by TLC and GC-MS analysis, the solvent was then removed, and the crude product was separated by column chromatography (eluted with petroleum ether/ethyl acetate = 6:1) to give a pure sample of **5a** (62%, 62 mg).

(*H*-Imidazo[1,2-*a*]pyridin-3-yl)(phenyl)methanone (3a).[11b] Yield: 89 mg, 80%; petroleum ether/ethyl acetate = 2:1 (R_f = 0.3). ^1H NMR (400 MHz, $CDCl_3$): δ 9.76 (d, J = 6.8 Hz, 1H), 8.22 (s, 1H), 7.89 (d, J = 7.2 Hz, 2H), 7.83 (d, J = 9.2 Hz, 1H), 7.61−7.52 (m, 4H), 7.17 (t, J = 7.2 Hz, 1H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 184.8, 149.0, 145.6, 139.2, 132.0, 129.4, 128.9, 128.4,, 123.4, 117.7, 115.1. MS (EI) m/z: 222, 193, 145, 117, 96, 90, 77, 51. Anal. Calcd for $C_{14}H_{10}N_2O$: C, 75.66; H, 4.54; N, 12.60. Found: C, 75.28; H, 4.51; N, 12.66.

(8-Methylimidazo[1,2-*a*]pyridin-3-yl)(phenyl)methanone (3b). Yield: 98 mg, 83%; white solid; mp 123.1 °C; petroleum ether/ethyl acetate = 3:1 (R_f = 0.28). ^1H NMR (400 MHz, $CDCl_3$): δ 9.61 (d, J = 6.8 Hz, 1H), 8.18 (s, 1H), 7.88 (d, J = 6.8 Hz, 2H), 7.62−7.51 (m, 3H), 7.37 (q, J = 7.6 Hz, 1H), 7.07 (t, J = 7.2 Hz, 1H), 2.70 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 184.7, 149.1, 144.9, 139.3, 131.8, 128.7, 128.5, 128.4, 127.5, 126.7, 126.5, 123.8, 115.0, 16.8. MS (EI) m/z: 236, 207, 159, 131, 104, 77. Anal. Calcd for $C_{15}H_{12}N_2O$: C, 76.25; H, 5.12; N, 11.86. Found: C, 75.86; H, 5.16; N, 11.93.

(7-Methylimidazo[1,2-*a*]pyridin-3-yl)(phenyl)methanone (3c).[11k] Yield: 99 mg, 84%; petroleum ether/ethyl acetate = 3:1 (R_f = 0.28). ^1H NMR (400 MHz, $CDCl_3$): δ 9.62 (d, J = 6.8 Hz, 1H), 8.15 (s, 1H), 7.87 (d, J = 7.2 Hz, 2H), 7.54−7.61 (m, 4H), 6.99 (d, J = 7.2 Hz, 1H), 2.51 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ184.4, 149.4, 145.8, 141.0, 139.2, 131.7, 128.6, 128.4, 127.9, 123.2, 117.5, 116.3, 21.5. MS (EI) m/z: 236, 207, 115, 77, 65. Anal. Calcd for $C_{15}H_{12}N_2O$: C, 76.25; H, 5.12; N, 11.86. Found: C, 75.96; H, 5.14; N, 11.90.

(6-Methyl-*H*-imidazo[1,2-*a*]pyridin-3-yl)(phenyl)methanone (3d). Yield: 98 mg, 80%; white solid; mp 125.5 °C; petroleum ether/ethyl acetate = 3:1 (R_f = 0.3). ^1H NMR (400 MHz, $CDCl_3$): δ 9.57 (s, 1H), 8.15 (s, 1H), 7.86 (d, J = 7.2 Hz, 2H), 7.72 (d, J = 9.2 Hz, 1H), 7.62 (m, 3H), 7.41 (s, 1H), 2.46 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 184.7, 148.0, 145.5, 139.4, 132.4, 131.9, 128.8, 128.5, 126.9, 126.8, 125.3, 116.9, 18.4. MS (EI) m/z: 236, 207, 159, 104, 77, 65. Anal. Calcd for $C_{15}H_{12}N_2O$: C, 76.25; H, 5.12; N, 11.86. Found: C, 75.89; H, 5.15; N, 11.91.

(5-Methylimidazo[1,2-*a*]pyridin-3-yl)(phenyl)methanone (3e). Yield: 91 mg, 77%; yellow oil; petroleum ether/ethyl acetate = 3:1 (R_f = 0.3). ^1H NMR (400 MHz, $CDCl_3$): δ 8.05 (d, J = 6.4 Hz, 3H), 7.70 (d, J = 8.8 Hz, 1H), 7.64 (t, J = 7.2 Hz, 1H), 7.54−7.45 (m, 3H), 6.91 (d, J = 7.2 Hz, 1H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 184.2, 150.7, 146.1, 139.8, 138.3, 132.7, 129.9, 129.2, 128.3, 125.7, 115.9, 115.0, 22.3. MS (EI) m/z: 236, 207, 159, 105, 77, 65. Anal. Calcd for $C_{15}H_{12}N_2O$: C, 76.25; H, 5.12; N, 11.86. Found: C, 75.89; H, 5.13; N, 11.91.

(6-Fluoro-*H*-imidazo[1,2-*a*]pyridin-3-yl)(phenyl)methanone (3f). Yield: 96 mg, 80%; yellow oil; petroleum ether/ethyl acetate = 3:1 (R_f = 0.28). ^1H NMR (400 MHz, $CDCl_3$): δ 9.77 (s, 1H), 8.26 (s,

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1H), 7.89 (d, J = 7.6 Hz, 2H), 7.83−7.91 (m, 1H), 7.64−7.46 (m, 4H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 184.9, 153.4, 138.9, 132.3, 128.9, 128.8, 128.7, 128.6, 121.1, 120.8, 118.0, 117.9, 116.5. MS (EI) m/z: 240, 211, 163, 135, 105, 77. Anal. Calcd for $C_{14}H_9FN_2O$: C, 69.99; H, 3.78; N, 11.66. Found: C, 69.64; H, 3.80; N, 11.71.

(6-Chloro-*H*-imidazo[1,2-*a*]pyridin-3-yl)(phenyl)methanone (3g). Yield: 110 mg, 85%; yellow oil; petroleum ether/ethyl acetate = 3:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.85 (d, J = 1.2 Hz, 1H), 8.22 (s, 1H), 7.90(d, J = 7.2 Hz, 2H), 7.77 (d, J = 9.2 Hz, 1H), 7.63 (d, J = 7.2 Hz, 1H), 7.56−7.51 (m, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ184.8, 156.7, 145.6, 138.8, 137.6, 132.3, 130.6, 128.8, 128.7, 126.9, 123.5, 118.0, 109.4.MS (EI) m/z: 258, 256, 181, 179, 151, 124, 105,77. Anal. Calcd for $C_{14}H_9ClN_2O$: C, 65.51; H, 3.53; N, 10.91. Found: C, 65.87; H, 3.50; N, 10.86.

(6-Bromo-*H*-imidazo[1,2-*a*]pyridin-3-yl)(phenyl)methanone (3h). Yield: 130 mg, 81%; yellow oil; petroleum ether/ethyl acetate = 3:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.94 (s, 1H), 8.19 (s, 1H), 7.88 (d, J = 7.2 Hz, 2H), 7.72 (d, J = 9.6 Hz, 1H), 7.63−7.61 (m, 2H), 7.56−7.53 (m, 2H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 184.8, 145.4, 138.9, 132.8, 132.3, 129.1, 128.8, 128.7, 118.3, 110.1, 77.4, 77.0, 76.7. MS (EI) m/z: 320, 301, 300, 225, 223, 116, 105, 96, 83, 77. Anal. Calcd for $C_{14}H_9BrN_2O$: C, 55.84; H, 3.01; N, 9.30. Found: C, 55.57; H, 3.02; N, 9.35.

(6-Iodoimidazo[1,2-*a*]pyridin-3-yl)(phenyl)methanone (3i). Yield: 139 mg, 80%; yellow oil; petroleum ether/ethyl acetate = 3:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 10.04 (s, 1H), 8.16 (s, 1H), 7.88 (d, J = 7.2 Hz, 2H), 7.75 (d, J = 8.8 Hz, 1H), 7.65−7.53 (m, 4H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 184.8, 145.0, 138.7, 137.5, 133.8, 132.4, 128.8, 128.7, 118.6, 100.0, 78.7. MS (EI) m/z: 348, 271, 245, 163, 105, 77. Anal. Calcd for $C_{14}H_9IN_2O$: C, 48.30; H, 2.61; N, 8.05. Found: C, 48.54; H, 2.59; N, 8.09.

(7-Chloroimidazo[1,2-*a*]pyridin-3-yl)(phenyl)methanone (3j). Yield: 107 mg, 83%; yellow oil; petroleum ether/ethyl acetate = 3:1 (R_f = 0.31). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.69 (d, J = 7.2 Hz, 1H), 8.20 (s, 1H), 7.88 (d, J = 7.2 Hz, 2H), 7.82 (s, 1H), 7.65−7.61 (m, 1H), 7.56−7.53 (m, 2H), 7.15 (d, J = 7.2 Hz, 1H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 184.8, 145.5, 138.8, 136.4, 132.4, 129.2, 128.8, 128.7, 116.8, MS (EI) m/z: 258, 256, 181, 179, 105, 77. Anal. Calcd for $C_{14}H_9IN_2O$: C, 48.30; H, 2.61; N, 8.05. Found: C, 48.54; H, 2.59; N, 8.10.

Phenyl(7-(trifluoromethyl)imidazo[1,2-*a*]pyridin-3-yl)-methanone (3k). Yield: 117 mg, 81%; yellow oil; petroleum ether/ethyl acetate = 4:1 (R_f = 0.29). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.87 (d, J = 6.4 Hz, 1H), 7.90 (d, J = 7.2 Hz, 2H), 7.69−7.41 (m, 6H), 7.31 (d, J = 7.2 Hz, 1H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 185.2, 146.1, 138.6, 132.6, 130.9, 130.8, 129.8, 129.7, 128.9, 128.8, 128.3, 127.4, 124.2, 121.4, 115.6, 110.8. MS (EI) m/z: 290, 261, 213, 185, 158, 138, 105, 77. Anal. Calcd for $C_{15}H_9F_3N_2O$: C, 62.07; H, 3.13; N, 9.65. Found: C, 61.78; H, 3.15; N, 9.71.

(6-Chloro-8-iodoimidazo[1,2-*a*]pyridin-3-yl)(phenyl)-methanone (3l). Yield: 145 mg, 76%; yellow oil; petroleum ether/ethyl acetate = 3:1 (R_f = 0.28). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.81 (s, 1H), 8.00 (s, 1H), 7.86 (d, J = 7.2 Hz, 2H), 7.65 (t, J = 7.2 Hz, 1H), 7.56 (t, J = 7.6 Hz, 2H), 7.39 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 185.1, 145.1, 138.9, 132.6, 129.1, 128.9, 128.8, 127.0, 126.7, 123.2, 84.0. MS (EI) m/z: 384, 382, 305, 279, 277, 135, 105, 77, 65. Anal. Calcd for $C_{14}H_8ClIN_2O$: C, 43.95; H, 2.11; N, 7.32. Found: C, 43.68; H, 2.13; N, 7.36.

(6,8-Diiodo-5-methylimidazo[1,2-*a*]pyridin-3-yl)(phenyl)-methanone (3m). Yield: 186 mg, 79%; yellow oil; petroleum ether/ethyl acetate = 3:1 (R_f = 0.32). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.34 (s, 1H), 8.04 (d, J = 7.6 Hz, 2H), 7.98 (s, 1H), 7.68 (t, J = 7.6 Hz, 1H), 7.57 (t, J = 7.6 Hz, 2H), 2.77 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 182.7, 149.3, 146.4, 145.2, 140.6, 137.6, 133.5, 130.2, 128.8, 128.5, 84.7, 81.5, 27.6. MS (EI) m/z: 488, 411, 383, 368, 226, 163, 135, 105, 77. Anal. Calcd for $C_{15}H_{10}I_2N_2O$: C, 36.91; H, 2.07; N, 5.74. Found: C, 36.72; H, 2.09; N, 5.79.

(8-Bromo-6-iodo-7-methylimidazo[1,2-*a*]pyridin-3-yl)-(phenyl)methanone(3n). Yield: 177 mg, 80%; yellow oil; petroleum ether/ethyl acetate = 3:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.90 (s, 1H), 7.87 (d, J = 7.6 Hz, 2H), 7.65 (t, J = 7.6 Hz, 1H), 7.56 (t, J = 7.6 Hz, 1H), 7.56 (t, J = 7.6 Hz, 2H), 7.40 (s, 1H), 2.84 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 185.0, 148.2, 143.8, 138.7, 132.5, 128.9, 128.8, 128.4, 111.6, 89.4, 29.0. MS (EI) m/z: 442, 440, 363, 335, 226, 208, 105, 77. Anal. Calcd for $C_{15}H_{10}BrIN_2O$: C, 40.85; H, 2.29; N, 6.35. Found: C, 40.64; H, 2.27; N, 6.40.

(6,8-Dibromo-7-methyl-*H*-imidazo[1,2-*a*]pyridin-3-yl)-(phenyl)methanone (3o). Yield: 163 mg, 82%; petroleum ether/ethyl acetate = 3:1 (R_f = 0.3); yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.92 (s, 1H), 7.87 (d, J = 7.6 Hz, 2H), 7.65 (t, J = 7.6 Hz, 1H), 7.56 (q, J = 7.6 Hz, 2H), 7.27 (s, 1H), 2.75 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ184.8, 147.1, 139.8, 138.7, 132.5, 129.1, 128.8, 127.7, 127.0, 113.0, 77.4, 77.0, 76.7, 23.4. MS (EI) m/z: 396, 394, 392, 318, 129, 105, 103, 77. Anal. Calcd for $C_{15}H_{10}Br_2N_2O$: C, 45.72; H, 2.56; N, 7.11. Found: C, 45.98; H, 2.54; N, 7.07.

1-(*H*-Imidazo[1,2-*a*]pyridin-3-yl)hexan-1-one (4a). Yield: 81 mg, 75%; yellow oil; petroleum ether/ethyl acetate = 3:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.66 (d, J = 6.8 Hz, 1H), 8.34 (s, 1H), 7.74 (d, J = 8.8 Hz, 1H), 7.48 (t, J = 7.6 Hz, 1H), 7.07 (t, J = 6.8 Hz, 1H), 2.91 (t, J = 7.2 Hz, 2H), 1.81−1.74 (m, 2H), 1.30−1.41 (m, 4H), 0.90 (t, J = 6.8 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 190.8, 142.8, 128.9, 128.8, 117.6, 115.0, 39.6, 31.6, 25.1, 22.5, 13.9. MS (EI) m/z: 216, 208, 180, 103, 43. Anal. Calcd for $C_{13}H_{16}N_2O$: C, 72.19; H, 7.46; N, 12.95. Found: C, 71.85; H, 7.48; N, 13.01.

1-(6-Methyl-*H*-imidazo[1,2-*a*]pyridin-3-yl)hexan-1-one (4b). Yield: 84 mg, 73%; yellow oil; petroleum ether/ethyl acetate = 3:1 (R_f = 0.32). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.5 (d, J = 6.8 Hz, 1H), 8.33 (s, 1H), 7.29 (d, J = 8.8 Hz, 1H), 7.00 (t, J = 6.8 Hz, 1H), 2.93 (t, J = 7.6 Hz, 2H), 2.67 (s, 3H), 1.71−1.82 (m, 2H), 1.35−1.41 (m, 4H), 0.93 (t, J = 6.8 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 190.8, 142.1, 127.9, 127.5, 126.6, 115.0, 39.6, 31.6, 25.1, 22.5, 16.9, 13.9. MS (EI) m/z: 230, 174, 159, 132, 104, 57. Anal. Calcd for $C_{14}H_{18}N_2O$: C, 73.01; H, 7.88; N, 12.16. Found: C, 72.79; H, 7.90; N, 12.22.

1-(7-Methyl-*H*-imidazo[1,2-*a*]pyridin-3-yl)hexan-1-one (4c). Yield: 91 mg, 79%; yellow oil; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.51 (d, J = 7.2 Hz, 1H), 8.28 (s, 1H), 7.49 (s, 1H), 6.89 (d, J = 7.2 Hz, 1H), 2.88 (t, J = 7.2 Hz, 2H), 2.45 (s, 3H), 1.78−1.73 (m, 2H), 1.34−1.38 (m, 4H), 0.91 (t, J = 6.4 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 190.5, 143.0, 140.5, 127.9, 117.4, 116.3, 39.4, 31.6, 25.1, 22.5, 21.5, 13.9. MS (EI) m/z: 230,187, 158, 132, 43. Anal. Calcd for $C_{14}H_{18}N_2O$: C, 73.01; H, 7.88; N, 12.16. Found: C, 72.77; H, 7.91; N, 12.23.

1-(6-Methyl-*H*-imidazo[1,2-*a*]pyridin-3-yl)hexan-1-one (4d). Yield: 87 mg, 76%; yellow oil; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.48 (s, 1H), 8.29 (s, 1H), 7.64 (d, J = 9.2 Hz, 1H), 7.32 (d, J = 8.4 Hz, 1H), 2.89 (t, J = 7.2 Hz, 2H), 2.39 (s, 3H), 1.73−1.81 (m, 2H), 1.35−1.41 (m, 4H), 0.91 (t, J = 0.89 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 190.7, 142.7, 131.8, 126.7, 125.0, 116.8, 39.5, 31.6, 25.1, 22.5, 18.4, 13.9. MS (EI) m/z: 230, 174, 159, 132, 104, 57. Anal. Calcd for $C_{14}H_{18}N_2O$: C, 73.01; H, 7.88; N, 12.16. Found: C, 72.81; H, 7.90; N, 12.21.

1-(5-Methyl-*H*-imidazo[1,2-*a*]pyridin-3-yl)hexan-1-one (4e). Yield: 81 mg, 70%; yellow oil; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.55 (d, J = 6.8 Hz, 1H), 8.33 (s, 1H), 7.30 (d, J = 9.6 Hz, 1H), 7.01 (t, J = 6.8 Hz, 1H), 2.94 (t, J = 7.6 Hz, 2H), 2.67 (s, 3H), 1.76−1.84 (m, 2H), 1.37−1.41 (m, 4H), 0.93 (t, J = 6.8 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 190.8, 142.2, 128.0, 127.5, 126.6, 115.0, 39.6, 31.6, 25.1, 22.5, 17.0, 14.0. MS (EI) m/z: 230, 187, 175, 174, 159, 133, 43. Anal. Calcd for $C_{14}H_{18}N_2O$: C, 73.01; H, 7.88; N, 12.16. Found: C, 72.85; H, 7.92; N, 12.22.

1-(6-Fluoroimidazo[1,2-*a*]pyridin-3-yl)hexan-1-one (4f). Yield: 87 mg, 74%; yellow oil; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.70 (q, J = 2.8 Hz, 1H), 8.37 (s, 1H), 7.76 (q, J = 5.2 Hz, 1H), 7.44−7.39 (m, 1H), 2.94 (t, J = 7.6 Hz, 2H), 1.84−1.77 (m, 2H), 1.41−1.34 (m, 4H), 0.94 (t, J = 6.8 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 190.9, 155.7, 153.3, 143.1, 120.5, 120.3, 117.9, 117.8, 116.2, 115.8, 39.6, 31.6, 24.9, 22.5, 13.9. MS (EI) m/z: 234, 207, 159, 131, 57. Anal. Calcd for $C_{13}H_{15}FN_2O$: C, 66.65; H, 6.45; N, 11.96. Found: C, 66.97; H, 6.42; N, 11.90.

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1-(6-Bromo-*H*-imidazo[1,2-*a*]pyridin-3-yl)hexan-1-one (4g). Yield: 112 mg, 76%; yellow oil; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.89 (s, 1H), 8.38 (t, J = 0.8 Hz, 1H), 7.68 (d, J = 9.2 Hz, 1H), 7.56 (d, J = 8.0 Hz, 1H), 2.93 (t, J = 7.2 Hz, 2H), 1.83−1.76 (m, 2H), 1.41−1.36 (m, 4H), 0.94 (t, J = 6.8 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 190.9, 142.6, 132.3, 129.0, 118.1, 110.0, 39.6, 31.5, 24.9, 22.5, 13.9. MS (EI) m/z: 296, 294, 223, 195, 132, 71, 43. Anal. Calcd for $C_{13}H_{15}BrN_2O$: C, 52.90; H, 5.12; N, 9.49. Found: C, 52.76; H, 5.14; N, 9.54.

1-(7-Chloroimidazo[1,2-*a*]pyridin-3-yl)hexan-1-one (4h). Yield: 94 mg, 75%; yellow oil; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.63 (d, J = 7.2 Hz, 1H), 8.35(s, 1H), 7.78 (d, J = 1.6 Hz, 1H), 7.07−7.09 (m, 1H), 2.93 (t, J = 7.2 Hz, 2H), 1.78−1.85 (m, 2H), 1.41−1.36 (m, 4H), 0.96 (t, J = 6.8 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 190.8, 143.2, 135.6, 129.0, 116.8, 116.4, 39.6, 31.6, 24.9, 22.5, 13.9. MS (EI) m/z: 252, 250, 236, 151, 43. Anal. Calcd for $C_{13}H_{15}ClN_2O$: C, 62.28; H, 6.03; N, 11.17. Found: C, 62.69; H, 5.99; N, 11.11.

1-(6,8-Dibromo-7-methyl-*H*-imidazo[1,2-*a*]pyridin-3-yl)-hexan-1-one (4i). Yield: 138 mg, 71%; yellow oil; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.86 (s, 1H), 8.30 (s, 1H), 2.92 (t, J = 7.2 Hz, 2H), 2.71 (s, 3H), 1.82−1.75 (m, 2H), 1.40−1.35 (m, 4H), 0.93 (t, J = 6.8 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 191.0, 142.5, 139.3, 127.6, 112.9, 112.6, 39.6, 31.5, 24.8, 23.3, 22.5, 13.9. MS (EI) m/z: 390, 388, 386, 371, 316, 236, 132, 57. Anal. Calcd for $C_{14}H_{16}Br_2N_2O$: C, 43.33; H, 4.16; N, 7.22. Found: C, 43.51; H, 4.14; N, 7.19.

1-(7-(Trifluoromethyl)imidazo[1,2-*a*]pyridin-3-yl)hexan-1-one (4j). Yield: 104 mg, 73%; yellow oil; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.80 (d, J = 7.2 Hz, 1H), 8.46 (s, 1H), 8.06 (s, 1H), 7.26 (d, J = 7.2 Hz, 1H), 2.98 (t, J = 7.6 Hz, 2H), 1.78−1.85 (m, 2H), 1.36−1.42 (m, 4H), 0.94 (t, J = 6.8 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 191.2, 143.4, 130.6, 130.2, 129.5, 126.8 (J = 276 Hz, 537 Hz), 115.5 (J = 4.4 Hz), 110.7, 110.6, 39.8, 31.5, 24.7, 22.4, 13.8. MS (EI) m/z: 284, 213, 185, 71, 43. Anal. Calcd for $C_{14}H_{15}F_3N_2O$: C, 59.15; H, 5.32; N, 9.85. Found: C, 59.47; H, 5.29; N, 9.81.

Imidazo[1,2-*a*]pyridine-3-carbaldehyde (4k).[11b,k] Yield: 51 mg, 70%; white solid; mp 113.2 °C; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.95 (s, 1H), 9.50 (d, J = 6.8 Hz, 1H), 8.34 (s, 1H), 7.82 (d, J = 8.8 Hz, 1H), 7.59 (t, J = 8.4 Hz, 1H), 7.17 (t, J = 6.2 Hz, 1H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 177.7, 149.2, 146.6, 130.0, 128.5, 124.9, 117.7, 115.4. MS (EI) m/z: 146, 117, 103, 90. Anal. Calcd for $C_8H_6N_2O$: C, 65.75; H, 4.14; N, 19.17. Found: C, 65.36; H, 4.17; N, 19.25;

8-Methylimidazo[1,2-*a*]pyridine-3-carbaldehyde (4l).[11b,k] Yield: 53 mg, 66%; white solid; mp 63.2 °C; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.92 (s, 1H), 9.35 (d, J = 6.8 Hz, 1H), 8.30 (s, 1H), 7.36 (d, J = 6.8 Hz, 1H), 7.05 (t, J = 6.8 Hz, 1H), 2.68 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 177.9, 149.5, 146.1, 129.1, 127.9, 126.4, 125.4, 115.5, 16.8. MS (EI) m/z: 160, 131, 104, 92, 65. Anal. Calcd for $C_9H_8N_2O$: C, 67.49; H, 5.03; N, 17.49. Found: C, 67.18; H, 5.06; N, 17.58.

6-Methylimidazo[1,2-*a*]pyridine-3-carbaldehyde (4m).[11b] Yield: 55 mg, 69%; white solid; mp 64.9 °C; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.93 (s, 1H), 9.33 (s, 1H), 8.31(s, 1H), 7.68 (d, J = 8.8 Hz, 1H), 7.41 (d, J = 8.8 Hz, 1H), (2.42)2.46 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 177.8, 148.4, 146.7, 133.0, 126.6, 125.7, 124.8, 116.9, 18.2. MS (EI) m/z: 160, 131, 104, 76, 51. Anal. Calcd for $C_9H_8N_2O$: C, 67.49; H, 5.03; N, 17.49. Found: C, 67.20; H, 5.03; N, 17.57.

7-Methylimidazo[1,2-*a*]pyridine-3-carbaldehyde (4n).[11k] Yield: 57 mg, 71%; white solid; mp 59.3 °C; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.86 (s, 1H), 9.34 (d, J = 6.8 Hz, 1H), 8.26 (s, 1H), 7.54 (s, 1H), 6.96 (d, J = 6.8 Hz, 1H), 2.48 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 177.4, 149.7, 147.0, 141.9, 127.8, 124.8, 117.8, 116.5, 21.6. MS (EI) m/z: 160, 131, 104, 65. Anal. Calcd for $C_9H_8N_2O$: C, 67.49; H, 5.03; N, 17.49. Found: C, 67.14; H, 5.06; N, 17.59.

Imidazo[1,2-*a*]pyrazin-3-yl(phenyl)methanone (4o). Yield: 58 mg, 52%; yellow oil; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.55 (d, J = 4.4 Hz, 1H), 9.34 (s, 1H), 8.33 (s, 1H), 8.28 (d, J = 4.4 Hz, 1H), 7.93 (d, J = 7.6 Hz, 2H), 7.69 (t, J = 7.2 Hz, 1H), 7.59 (t, J = 7.6 Hz, 2H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 185.4, 144.8, 144.1, 138.3, 132.9, 132.7, 128.9, 128.8, 121.0, 120.9. MS (EI) m/z: 223, 146, 118, 105, 77. Anal. Calcd for $C_{13}H_9N_3O$: C, 69.95; H, 4.06; N, 18.82. Found: C, 69.64; H, 4.09; N, 19.01.

1-(Imidazo[1,2-*a*]pyrazin-3-yl)hexan-1-one (4p). Yield: 49 mg, 45%; yellow oil; petroleum ether/ethyl acetate = 4:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.48 (dd, J = 1.2, 4.8 Hz, 1H), 9.28 (d, J = 1.2 Hz, 1H), 8.43 (s, 1H), 8.20 (d, J = 4.8 Hz, 1H), 2.99 (t, J = 7.6 Hz, 2H), 1.78−1.85 (m, 2H), 1.38−1.42 (m, 4H), 0.94 (t, J = 6.8 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 191.6, 144.0, 142.3, 132.5, 120.8, 40.0, 31.4, 24.5, 22.4, 13.9. MS (EI) m/z: 217, 189, 163, 120, 71, 43. Anal. Calcd for $C_{12}H_{15}N_3O$: C, 66.34; H, 6.96; N, 19.34. Found: C, 66.01; H, 7.01; N, 19.52;

3-((*E*)-Hex-1-enyl)*H*-imidazo[1,2-*a*]pyridine (5a). Yield: 62 mg, 62%; yellow oil; petroleum ether/ethyl acetate = 7:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.10 (d, J = 6.8 Hz, 1H), 7.68 (s, 1H), 7.63 (d, J = 9.2 Hz, 1H), 7.17 (t, J = 7.6 Hz, 1H), 6.85 (t, J = 6.8 Hz, 1H), 6.47 (d, J = 16 Hz, 1H), 6.29−6.24(m, 1H), 2.32 (q, J = 7.2 Hz, 2H), 1.54−1.36 (m, 4H), 0.97 (t, J = 7.2 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 132.9, 130.7, 130.6, 123.7, 123.2, 118.0, 114.8, 112.5, 33.2, 31.5, 22.3, 13.9. MS (EI) m/z: 200, 143, 117, 43. Anal. Calcd for $C_{13}H_{16}N_2$: C, 77.96; H, 8.05; N, 13.99. Found: C, 77.68; H, 8.09; N, 13.88.

3-((*E*)-Hex-1-enyl)-5-methyl-*H*-imidazo[1,2-*a*]pyridine (5b). Yield: 59 mg, 54%; yellow oil; petroleum ether/ethyl acetate = 6:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.56−7.47 (m, 2H), 7.05 (t, J = 7.2 Hz, 1H), 6.82 (d, J = 15.6 Hz, 1H), 6.52 (d, J = 6.8 Hz, 1H), 6.04−5.97 (m, 1H), 2.82 (s, 3H), 2.26 (q, J = 6.8 Hz, 2H), 1.53−1.34 (m, 4H), 0.96 (t, J = 7.2 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 136.5, 136.4, 126.7, 118.8, 115.9, 113.6, 113.5, 32.8, 31.2, 22.3, 21.4, 13.9. MS (EI) m/z: 214, 171, 155, 133, 92, 65. Anal. Calcd for $C_{14}H_{18}N_2$: C, 78.46; H, 8.47; N, 13.07. Found: C, 78.08; H, 8.51; N, 13.12.

3-((*E*)-Hex-1-enyl)-7-methyl-*H*-imidazo[1,2-*a*]pyridine (5c). Yield: 72 mg, 67%; yellow oil; petroleum ether/ethyl acetate = 6:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.99 (d, J = 6.8 Hz, 1H), 7. 60 (s, 1H), 7.39 (s, 1H), 6.68 (d, J = 6.4 Hz, 1H), 6.44 (d, J = 15.6 Hz, 1H), 6.25−6.17 (m, 1H), 2.39 (s, 3H), 2.31 (q, J = 6.8 Hz, 2H), 1.53−1.35 (m, 4H), 0.96 (t, J = 7.6 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 145.8, 132.2, 132.2, 122.5, 121.4, 116.3, 115.2, 114.9, 33.2, 31.5, 22.3, 21.2, 13.9. MS (EI) m/z: 214, 171, 159, 132, 43. Anal. Calcd for $C_{14}H_{18}N_2$: C, 78.46; H, 8.47; N, 13.07. Found: C, 78.07; H, 8.51; N, 13.13.

3-((*E*)-Hex-1-enyl)-8-methyl-*H*-imidazo[1,2-*a*]pyridine (5d). Yield: 67 mg, 63%; yellow oil; petroleum ether/ethyl acetate = 6:1 (R_f = 0.3). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.96 (d, J = 6.8 Hz, 1H), 7.65 (s, 1H), 6.94 (d, J = 6.8 Hz, 1H), 6.75 (t, J = 6.8 Hz, 1H), 6.45 (d, J = 16 Hz, 1H), 6.27−6.20 (m, 1H), 2.60 (s, 3H), 2.31 (q, J = 6.8 Hz, 2H), 1.53−1.35 (m, 4H), 0.96 (t, J = 7.2 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 142.1, 132.5, 129.9, 122.6, 121.1, 115.1, 112.4, 33.2, 31.4, 22.3, 17.0, 13.9. MS (EI) m/z: 214, 185, 158, 132, 43. Anal. Calcd for $C_{14}H_{18}N_2$: C, 78.46; H, 8.47; N, 13.07. Found: C, 78.09; H, 8.52; N, 13.14.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

^{1}H and ^{13}C NMR spectra. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: caohua@gdpu.edu.cn.

Notes

The authors declare no competing financial interest.

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