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I_2-Catalyzed Three-Component Consecutive Reaction for the Synthesis of 3-Aroylimidazo[1,2-*a*]-*N*-Heterocycles

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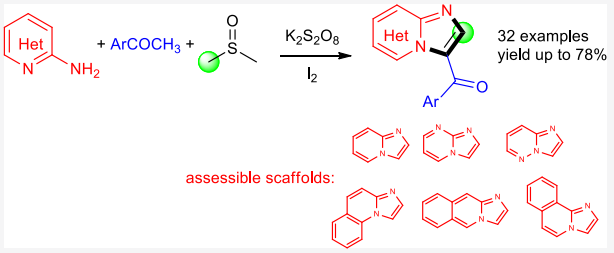
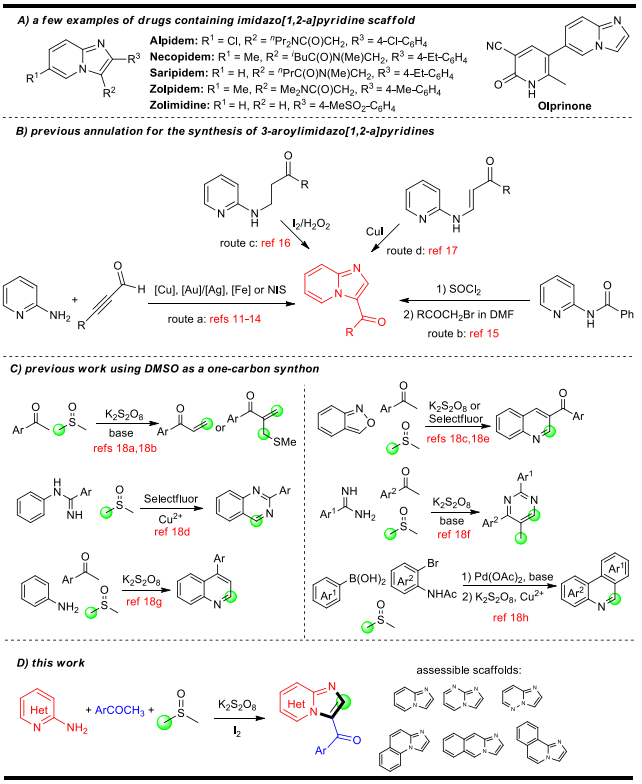
ABSTRACT: A convenient one-pot, three-component reaction has been developed for the synthesis of 3-aroylimidazo[1,2-*a*]-*N*-heterocycles from aryl ketones and 2-amino-*N*-heterocycles using dimethyl sulfoxide as a methylene donor. The reaction proceeds smoothly catalyzed by I_2 in the presence of $K_2S_2O_8$ and affords the desired products in moderate to good yields. This protocol offers significant superiority in accessing biologically active 3-aroylimidazo[1,2-*a*]-*N*-heterocycles with various substitution patterns.



INTRODUCTION

Nitrogen-containing heterocycles have been widely found in pharmaceutical chemistry, natural products, and materials science.[1] Imidazo[1,2-*a*]pyridines are aza-fused heterocyclic compounds and they exhibit a wide distribution in many pharmacologically important compounds as well as in the field of materials and organometallic chemistry.[2] For example, the clinically used drugs alpidem,[3] necopidem,[4] saripidem[4] (all used as an anxiolytic agent), olprinone (for treatment of acute heart failure),[5] zolpidem (for treatment of insomnia),[3] and zolimidine (for treatment of peptic ulcer)[6] have imidazo[1,2-*a*]pyridine scaffolds (Scheme 1A). Accordingly, there are a lot of well-documented methods for the synthesis of imidazo[1,2-*a*]pyridines established over the past few decades.[7] These can be classified based on the reaction type into condensation, tandem reaction, multicomponent reaction, oxidative coupling, aminooxygenation, and hydroaminoation.[8] However, only a few reports demonstrate the synthesis of 3-aroylimidazo[1,2-*a*]pyridines. This type of compound is also an important substructure in various drug candidates such as for calcium channel blockers[9] and anticancer activities.[10] It can be prepared directly from aminooxygenation of 2-aminopyridines with 3-arylpropiolaldehydes catalyzed by Fe,[11] Au/Ag,[12] Cu,[13] or NIS[14] (Scheme 1B, route a). In 2012, Schmitt et al. reported a cascade synthesis of 3-aroylimidazo[1,2-*a*]pyridines from *N*-acylamidine with thionyl chloride followed by a reaction with phenacyl bromide in DMF (Scheme 1B, route b).[15] Recently, He's group developed intramolecular oxidative α-amination of carbonyl compounds by I_2/H_2O_2 to give 3-aroylimidazo[1,2-*a*]pyridines (Scheme 1B, route c).[16] Similarly, Wen et al. demonstrated copper-catalyzed intramolecular oxidative amination of enaminones for the synthesis of 3-aroylimidazo[1,2-*a*]pyridines (Scheme 1B, route d).[17] Despite these findings, developing a convenient method is still desirable in hit-to-lead drug discovery, which requires quick

Scheme 1. Drug-Based Imidazo[1,2-*a*]pyridines, Previous Reports and Our Design.



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follow-up to construct 3-aroylimidazo[1,2-*a*]pyridines from readily accessible and inexpensive starting materials in a single operation under mild conditions. Continuing our efforts to develop methods for constructing heterocycles and inspired by previous reports that used dimethyl sulfoxide (DMSO) as a one-carbon synthon (Scheme 1C),[18] here we would like to describe the one-pot synthesis of 3-aroylimidazo[1,2-*a*]-*N*-heterocycles from aryl ketones and 2-amino-*N*-heterocycles using the DMSO/$K_2S_2O_8$ system catalyzed by I_2 (Scheme 1D).

RESULTS AND DISCUSSION

Initially, for a clear structure elucidation, we chose 2-aminopyridine (**1a**) and 1-(*p*-tolyl)ethanone (**2b**) as a model to investigate the optimal reaction conditions. The effects of different oxidants, the amount of the oxidant, different additives, different ratios of the substrates, the amount of I_2, and the reaction temperatures on the product yields were investigated. After tedious optimization experiments (see Supporting Information, Tables S1−S4), $K_2S_2O_8$ (3 equiv) was selected as the oxidant. In the presence of I_2 (0.1 equiv), the desired 3-aroylimidazo[1,2-*a*]pyridine (**3ab**) was obtained in 20% yield (Table 1, entry 1). No product was obtained in

Table 1. Optimization of Reaction Conditions[a]

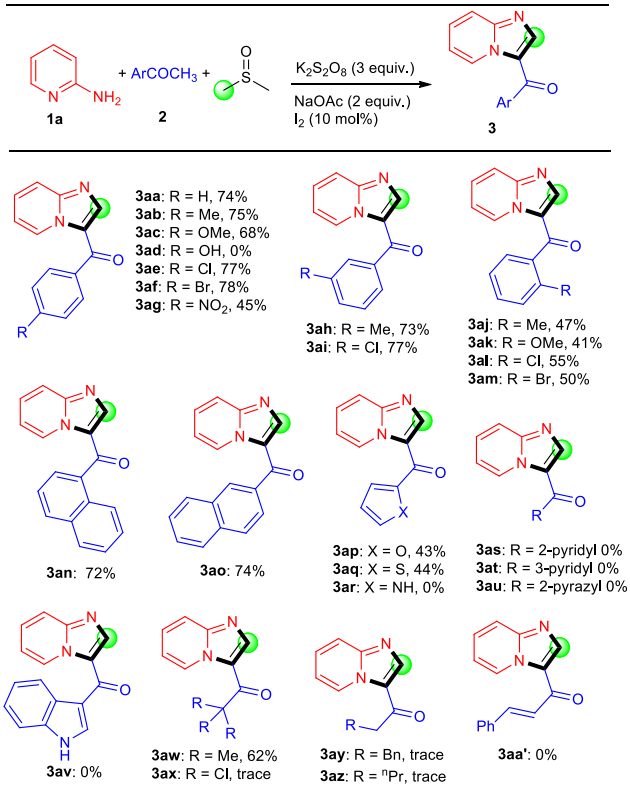
entry	ratio of 1a:2b	NaOAc (equiv)	I_2 (mmol)	*T* (°C)	yield (%)[b]
1	1:1.2	0	0.1	120	20
2	1:1.2	0	0	120	0
3	1:1.2	0	1	120	0
4[c]	1:1.2	2	0.1	120	33
5[d]	1:1.2	2	0.1	120	40
6[d]	1:1.2	3	0.1	120	26
7[d]	1:1.2	2	0.1	130	22
8[d]	**1:1.2**	**2**	**0.1**	**110**	**75**
9[d]	1:1.2	2	0.1	100	53
10[d]	1:1.2	2	0.1	90	39
11[d]	1:1	2	0.1	110	61
12[d]	1:0.8	2	0.1	110	41

[a]Reaction conditions: 2-aminopyridine (**1a**, 1 mmol), 1-(*p*-tolyl)-ethanone (**2b**, 1.2 mmol), $K_2S_2O_8$ (3 mmol), NaOAc (2 mmol), and I_2 at the indicated amount in DMSO (2 mL) were heated at the indicated temperature for 16 h. [b]Isolated yield. [c]**1a** and I_2 were added after 9 h and then the mixture was heated for another 7 h. [d]I_2 was added after 9 h and then the mixture was heated for another 7 h.

the absence of I_2, indicating that I_2 is necessary for this transformation (entry 2). On the other hand, when a stoichiometric amount of I_2 was added to the reaction, although the starting materials were completely converted, the expected product was not detected, which may be due to overoxidation (entry 3). The efficiency of this conversion was improved by adding NaOAc (2 mmol) to the reaction mixture, and the product yield increased from 20 to 33% when **1a** and I_2 were added 9 h later (entry 4). A further 7% increase was achieved by changing the feeding order (entry 5). Increasing the amount of NaOAc to 3 mmol led to a lower yield of **3ab** (entry 6). The reaction temperature was then investigated and after brief screening of the different temperatures, we found that the highest yield was achieved at 110 °C (entries 7−10). With the ratio of the two substrates screened, the best result was obtained when 1:1.2 of **1a**:**2b** was used (entry 8 vs entries 11 and 12). Based on these studies, the optimal reaction conditions were established as listed in entry 8.

Having established the optimal reaction conditions, we began to investigate the substrate scope and the generality of the reaction with respect to various aryl methyl ketones with 2-aminopyridines. As shown in Table 2, a series of *para*-, *meta*-, and *ortho*-substituted acetophenones bearing electron-donating groups (e.g., −Me and −OMe) and electron-withdrawing groups (e.g., −Cl, −Br, and $-NO_2$) was smoothly transformed into the respective 3-aroylimidazo[1,2-*a*]pyridines in moderate to good yields (**3aa**−**3am**). These results also indicated that aryl ketones bearing electron-withdrawing groups provided slightly higher yields in comparison with those bearing electron-donating groups (**3ae** and **3af** vs **3ab** and **3ac**, **3ai** vs **3ah**, and **3al** and **3am** vs **3aj** and **3ak**). However, *para*-NO_2-substituted acetophenone (**2g**) gave **3ag** in a moderate yield only. No corresponding product was observed with *para*-hydroxyl acetophenone (**2d**) as the substrate. On the other hand, the position of the substituent group on the phenyl ring significantly influenced the yield. For instance, *para*- and *meta*-methyl-substituted acetophenones gave the corresponding products at 75% (**3ab**) and 73% (**3ah**) yields, respectively, while the *ortho*-methyl analogue proceeded in the reaction with 47% (**3aj**) yield only. A similar phenomenon was observed for

Table 2. Substrate Scope Based on Aryl Ketones 2[a,b]



[a]Reaction conditions: **1a** (1 mmol), **2** (1.2 mmol), $K_2S_2O_8$ (3 mmol), and NaOAc (2 mmol) in DMSO (2 mL) were heated at 110 °C for 9 h followed by addition of I_2 (0.1 mmol) and the mixture was heated for another 7 h. [b]Isolated yield.

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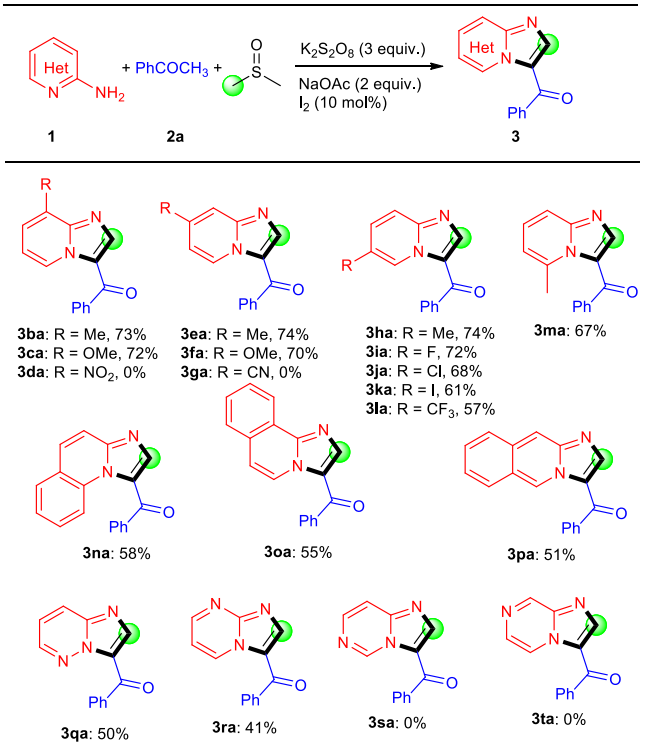
chloro-substituted analogues (**3ae** and **3ai** vs **3al**). In addition, naphthyl and heterocyclic ketones were also investigated for this transformation. The corresponding imidazo[1,2-*a*]-pyridines were afforded in good yields for 1- and 2-naphthyl substituents (**3an** and **3ao**) and moderate yields for 2-furyl and 2-thienyl (**3ap** and **3aq**), but the transformation failed with 2-pyrrolyl (**3ar**), 2-pyridyl (**3at**), 3-pyridyl (**3au**), and 2-pyrazyl (**3av**) substituents. It is likely that the *N*-heteroaryl ketones underwent oxidation in the presence of $K_2S_2O_8$ to form a N−O bond and therefore no desired products were obtained. Furthermore, aliphatic ketones such as pinacolone (**2w**), 1,1,1-trichloroacetone (**2x**), 4-phenyl-2-butanone (**2y**), and 2-hexanone (**2z**) were also attempted for this transformation, but only pinacolone gave the respective product **3at** in a moderate yield. This may be due to the effect of the electron-withdrawing property of the CCl_3 group (**2x**) or the existence of two active *α*-H on both sides of the ketone group (**2y** and **2z**). 4-Phenyl-3-buten-2-one (**2a'**) was also not suitable for this transformation and the formation of the desired product (**3aa'**) was unsuccessful. It should be noted that when **2r**−**2v** and **2w**−**2a'** were reacted with **1a** under the standard conditions before the addition of I_2, the starting materials were consumed completely but only unidentified complexes were detected by thin-layer chromatography (TLC). The addition of I_2 after 9 h did not give the target molecules.

To further explore the universality of this three-component annulation reaction, several heteroaryl amines **1** were reacted with acetophenone **2a** under the optimal reaction conditions (Table 3). We first examined various substituents on different positions of the 2-amino pyridine ring. The result showed that all of the transformations except for **1d** and **1g** were well tolerated and the corresponding imidazo[1,2-*a*]pyridines (**3ba**−**3ma**) were afforded in good yields. The position of the substituent had little influence on the reaction efficiency. In addition, 2-aminoquinoline (**1n**), 1-aminoisoquinoline (**1o**), and 3-aminoisoquinoline (**1p**) were also studied for the transformation and moderate yields (51−58%) of the corresponding imidazo[1,2-*a*]pyridines were achieved. To further expand the substrate scope, other four heteroaryl amines were investigated for the conversion: 3-amino-pyridazine (**1q**) and 2-aminopyrimidine (**1r**) were found to be smoothly converted into the corresponding imidazo[1,2-*a*]pyridines (**3qa** and **3ra**, respectively) in moderate yields; however, to our surprise, 4-aminopyrimidine (**1s**) and aminopyrazine (**1t**) failed to undergo this conversion. Such reactions were repeated twice but the same results were achieved. Similar to the aforesaid unsuccessful reactions illustrated in Table 2, the substrates were consumed completely (monitored by TLC) but no desired intermediates were afforded before the addition of I_2 under such reaction conditions.

To assess the efficiency and potential for application of this method, we employed this method for the synthesis of (7-methoxyimidazo[1,2-*a*]pyridine-3-yl)(3,4,5-trimethoxy-phenyl)methanone (**3fb'**), an anticancer drug candidate.[10] Hsieh's group reported the synthesis of **3fb'** from 2-amino-4-methoxypyridine (**1f**) via a three-step reaction with a total yield of 20%. In our case, a one-pot synthesis of **3fb'** with a yield of 61% has been developed successfully from commercially available substrates, which has apparent superiority to the literature report (Scheme 2).

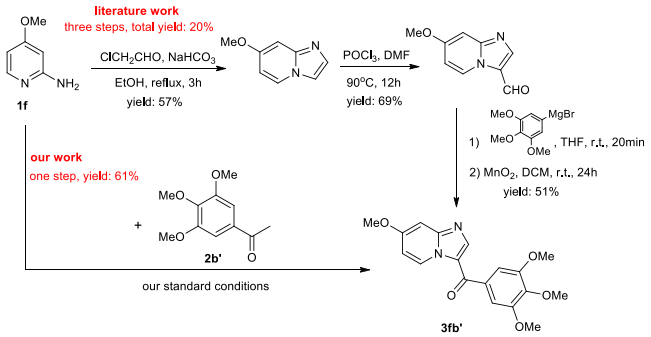
To gain insight into the reaction mechanism, several control experiments were carried out (Scheme 3). 1-(*p*-Tolyl)-

Table 3. Substrate Scope Based on Heterocyclic Amines 1[a,b]



[a]Reaction conditions: **1** (1 mmol), **2a** (1.2 mmol), $K_2S_2O_8$ (3 mmol), and NaOAc (2 mmol) in DMSO (2 mL) were heated at 110 °C for 9 h followed by addition of I_2 (0.1 mmol) and the mixture was heated for another 7 h. [b]Isolated yield.

Scheme 2. Comparison of Our Method with the Literature Report for the Synthesis of 3fb'.



ethenone (**2b**) has been reported to react with DMSO oxidized by $K_2S_2O_8$ to generate *α*,*β*-enone (**4b**).[18a]-c In the presence of 2-aminopyridine, the intermediate **4b** can undergo Michael addition to generate compound **5ab**, a key intermediate of the imidazo[1,2-*a*]pyridine (Scheme 3, eq 1). On the other hand, the reaction of 2-aminopyridine with DMSO was carried out in the presence of $K_2S_2O_8$ and the generated intermediate (**6a**) was subsequently coupled with **2b** to form compound **5ab** (Scheme 3, eq 2). The compound **5ab** was then catalyzed by I_2 in DMSO to afford the desired product **3ab** (Scheme 3, eq 3). A kinetic deuterium isotope study showed that a **3ab**/**3ab'** ratio of 1.86 was achieved when the reaction with an equal amount of DMSO/DMSO-d_6 was tested under the optimal conditions (Scheme 3, eq 4),

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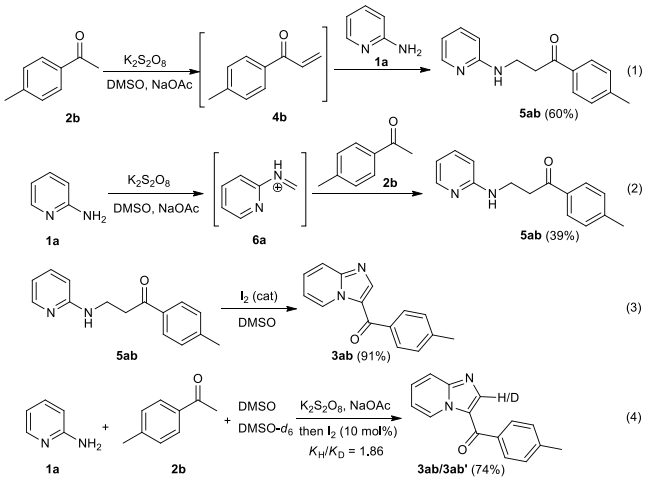
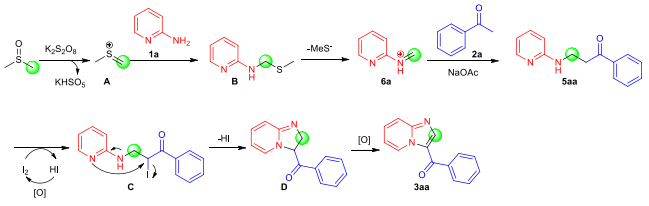
Scheme 3. Control Experiments.



indicating that the C(sp^3)−H bond cleavage of DMSO might be involved in the rate-limiting step.

Although a more detailed study might be needed to fully clarify the reaction mechanism, a plausible pathway is proposed based on the above experimental results and literature reports[18a,19] (Scheme 4). Results from Scheme 3

Scheme 4. Possible Reaction Mechanism.



indicate that both pathways via the intermediates **4b** and **6a** are possible. However, we found that in the optimal reaction conditions, 2-aminopyridine (**1a**) was consumed much faster than acetophenone, indicating that the pathway via the intermediate **6a** is more likely to form. On the other hand, compared to acetophenone, the higher nucleophilicity of the amine from 2-aminopyridine (**1a**) renders it easier to react with the sulfenium ion (**A**). A coupling of the intermediate **6a** with **2a** in the presence of NaOAc is performed and the resulting compound **5aa** is reacted with I_2 to afford intermediate **C** and HI, which is reoxidized to complete the catalytic cycle. The intermediate **C** experiences a similar S_N^2 process to form a cyclic intermediate **D**. The nonaromatic intermediate **D** is easily oxidized to generate the final target product **3aa**.

CONCLUSIONS

In conclusion, we have developed an efficient one-pot consecutive protocol for the construction of 3-aroylimidazo-[1,2-*a*]-*N*-heterocycles via *α*-C(sp^3)−H methylenation of ketones, coupling with 2-amino-*N*-heterocycles, iodination, annulation, and oxidation processes. Moderate to good yields of the products were achieved using the $I_2/K_2S_2O_8$ system. This protocol offers significant advantages for accessing biologically active imidazopyridine derivatives. Further studies of DMSO as the methylene source for annulation reactions to construct heterocycles are underway in our laboratory.

EXPERIMENTAL SECTION

General Methods. All reagents and solvents were obtained from commercial sources unless otherwise stated. All raw materials were obtained from commercial sources. All experiments were conducted in the air. All the reactions were monitored by TLC. TLC was performed on precoated silica gel plates (Qingdao Haiyang Chemical Co., Ltd., China). Column chromatography was performed on silica gel (240−400 mesh) with petroleum ether and ethyl acetate as eluents. ^{1}H and ^{13}C nuclear magnetic resonance (NMR) (400 and 101 MHz) spectra were recorded using a Bruker Avance 400 MHz using $CDCl_3$ as a solvent with tetramethylsilane as the internal standard. Melting points were determined using a WRS-1B apparatus and were uncorrected. High-resolution mass spectra were obtained with the Q-TOF-Premier mass spectrometer. Reactions were monitored by TLC and visualized with ultraviolet light.

General Procedure for the Synthesis of 3 (3ab as an Example). The reaction was carried out with 2-aminopyridine (**1a**, 94 mg, 1 mmol), 1-(*p*-tolyl)ethanone (**2b**, 161 mg, 1.2 mmol), $K_2S_2O_8$ (810 mg, 3 mmol), and NaOAc (164 mg, 2 mmol) in DMSO (2 mL) heated at 110 °C in an oil bath for 9 h. I_2 (25.3 mg, 0.1 mmol) was added, and then the mixture was heated in the oil bath for another 7 h. The reaction was monitored by TLC. Once the reaction was complete, the reaction mixture was treated with H_2O (15.0 mL) and CH_2Cl_2 (8.0 mL). The organic and aqueous layers were then separated, and the aqueous layer was extracted with CH_2Cl_2 (3 × 8 mL). The combined organic extracts were dried (Na_2SO_4), then the solvent was removed under a reduced pressure, and the remaining residue was purified by column chromatography. Compound **3ab** (178 mg, 75% yield) was obtained as a white solid.

*Imidazo[1,2-a]pyridin-3-yl(phenyl)methanone (**3aa**).*[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 164 mg, 74%); mp 96−97 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.76 (d, J = 9.2 Hz, 1H), 8.22 (s, 1H), 7.89 (d, J = 6.8 Hz, 2H), 7.82 (d, J = 9.2 Hz, 1H), 7.62 (t, J = 7.2 Hz, 1H), 7.58−7.52 (m, 3H), 7.16 (t, J = 6.8 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184. 9, 149.1, 145.7, 139.3, 132.1, 129.5, 128.9, 128.9, 128.6, 123.6, 117.8, 115.2. ESI-MS: m/z $[M + H]^+$ 223.

*Imidazo[1,2-a]pyridin-3-yl(p-tolyl)methanone (**3ab**).*[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 177 mg, 75%); mp 109−100 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.74 (d, J = 7.2 Hz, 1H), 8.22 (s, 1H), 7.82−7.79 (m, 3H), 7.55 (t, J = 6.8 Hz, 1H), 7.34 (d, J = 7.6 Hz, 2H), 7.15 (t, J = 6.8 Hz, 1H), 2.47 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184.7, 149.0, 145.3, 142.8, 136.6, 129.3, 129.3, 129.0, 128.9, 123.6, 117.7, 115.0, 21.6. ESI-MS: m/z $[M + H]^+$ 237.

*Imidazo[1,2-a]pyridin-3-yl(4-methoxyphenyl)methanone (**3ac**).*[17] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 171 mg, 68%); mp 140−141 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.70 (d, J = 6.8 Hz, 1H), 8.21 (s, 1H), 7.91 (t, J = 8.4 Hz, 2H), 7.80 (d, J = 8.8 Hz, 1H), 7.53 (t, J = 8.8 Hz, 1H), 7.13 (t, J = 6.8 Hz, 1H), 7.04 (d, J = 8.8 Hz, 2H), 3.91 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 183.8, 163.0, 148.9, 144.8, 131.9, 131.1, 129.1, 128.9, 123.6, 117.7, 114.9, 113.9, 55.5. ESI-MS: m/z $[M + H]^+$ 253.

*(4-Chlorophenyl)(imidazo[1,2-a]pyridin-3-yl)methanone (**3ae**).*[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 197 mg, 77%); mp 192−193 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.72 (d, J = 6.8 Hz, 1H), 8.19 (s, 1H), 7.84−7.81 (m, 3H), 7.57 (t, J = 8.0 Hz, 1H), 7.52 (d, J = 8.4 Hz, 2H), 7.17 (t, J = 8.0 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 183.4, 149.3, 145.6, 138.4, 137.6, 130. 2, 129.7, 129.0, 128.9, 123.4, 117.9, 115.3. ESI-MS: m/z $[M + H]^+$ 257.

*(4-Bromophenyl)(imidazo[1,2-a]pyridin-3-yl)methanone (**3af**).*[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 199 mg, 78%); mp 204−205 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.73 (d, J = 5.6 Hz, 1H), 8.19 (s, 1H), 7.82 (d, J = 9.2 Hz, 1H), 7.76 (d, J = 8.0 Hz, 2H), 7.69 (d, J = 8.4 Hz, 2H), 7.58 (t, J = 7.2 Hz, 1H), 7.17 (t, J = 4.0 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 183.5, 149.3, 145.6, 138.0, 131.9, 130.4, 129.7, 128.9, 126.9, 123.3, 117.9, 115.3. ESI-MS: m/z $[M + H]^+$ 301.

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Imidazo[1,2-a]pyridin-3-yl(4-nitrophenyl)methanone (**3ag**).[14] Petroleum ether/ethyl acetate = 1:1; yellow powder (yield: 134 mg, 45%); mp 184−185 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.76 (d, J = 6.8 Hz, 1H), 8.41 (d, J = 8.8 Hz, 2H), 8.19 (s, 1H), 8.03 (d, J = 8.8 Hz, 2H), 7.87 (d, J = 8.8 Hz, 1H), 7.64 (t, J = 7.2 Hz, 1H), 7.23 (d, J = 6.8 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 171.2, 146.2, 144.5, 144.3, 130.4, 130.3, 129.7, 129.0, 123.9, 118.0, 115.8, 115.5. ESI-MS: m/z [M + H]$^+$ 268.

Imidazo[1,2-a]pyridin-3-yl(m-tolyl)methanone (**3ah**).[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 172 mg, 73%); mp 123−124 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.75 (d, J = 6.0 Hz, 1H), 8.21 (s, 1H), 7.81 (d, J = 8.8 Hz, 1H), 7.69−7.66 (m, 2H), 7.55 (t, J = 6.4 Hz, 1H), 7.43 (d, J = 4.8 Hz, 2H), 7.16 (t, J = 7.2 Hz, 1H), 2.46 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 185.1, 149.1, 145.7, 139.3, 138.5, 132.8, 129.4, 129.4, 128.9, 128.4, 126.1, 123.6, 117.8, 115.1, 21.4. ESI-MS: m/z [M + H]$^+$ 237.

(3-Chlorophenyl)(imidazo[1,2-a]pyridin-3-yl)methanone (**3ai**). Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 197 mg, 77%); mp 180−181 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.73 (d, J = 6.8 Hz, 1H), 8.21 (s, 1H), 7.85 (s, 1H), 7.84 (d, J = 9.2 Hz, 1H), 7.75 (d, J = 7.6 Hz, 1H), 7.60−7.56 (m, 2H), 7.48 (t, J = 8.0 Hz, 1H), 7.18 (t, J = 6.8 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 183.1, 149.4, 145.9, 140.9, 134.9, 132.0, 129.9, 129.8, 128.93, 128.9, 126.9, 123.3, 117.9, 115.4. HRMS (ESI): calcd for $C_{14}H_{10}ClN_2O$ m/z [M + H]$^+$ 257.0476; found 257.0480.

Imidazo[1,2-a]pyridin-3-yl(o-tolyl)methanone (**3aj**). Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 110 mg, 47%); mp 108−109 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.82 (d, J = 6.8 Hz, 1H), 7.93 (s, 1H), 7.81 (d, J = 9.2 Hz, 1H), 7.57 (d, J = 8.8 Hz, 1H), 7.48 (d, J = 7.2 Hz, 1H), 7.42 (t, J = 6.8 Hz, 1H), 7.32 (t, J = 7.2 Hz, 2H), 7.17 (t, J = 6.8 Hz, 1H), 2.43 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 186.9, 149.3, 146.5, 139.0, 136.5, 131.2, 130.3, 129.6, 129.0, 128.3, 125.3, 124.5, 117.8, 115.3, 19.6. HRMS (ESI): calcd for $C_{15}H_{13}N_2O$ m/z [M + H]$^+$ 237.1022; found 237.1026.

Imidazo[1,2-a]pyridin-3-yl(2-methoxyphenyl)methanone (**3ak**). Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 100 mg, 41%); mp 110−111 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.81 (d, J = 6.8 Hz, 1H), 7.96 (s, 1H), 7.79 (d, J = 9.2 Hz, 1H), 7.55 (t, J = 7.2 Hz, 1H), 7.51−7.44 (m, 2H), 7.16 (t, J = 6.8 Hz, 1H), 7.06 (m, 2H), 3.83 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184.3, 157.0, 146.4, 131.9, 129.4, 129.4, 129.1, 129.0, 124.6, 120.4, 117.7, 115.2, 111.6, 55.7. HRMS (ESI): calcd for $C_{15}H_{13}N_2O_2$ m/z [M + H]$^+$ 253.0972; found 253.0975.

(2-Chlorophenyl)(imidazo[1,2-a]pyridin-3-yl)methanone (**3al**). Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 140 mg, 55%); mp 154−155 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.79 (d, J = 6.8 Hz, 1H), 7.92 (s, 1H), 7.82 (d, J = 8.8 Hz, 1H), 7.60 (t, J = 7.2 Hz, 1H), 7.57−7.50 (m, 2H), 7.47 (t, J = 7.6 Hz, 1H), 7.40 (t, J = 7.6 Hz, 1H), 7.20 (t, J = 6.8 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 183.0, 149.9, 146.9, 138.5, 131.5, 131.3, 130.4, 130.0, 129.2, 129.0, 126.7, 123.9, 117.9, 115.6. HRMS (ESI): calcd for $C_{14}H_{10}ClN_2O$ m/z [M + H]$^+$ 257.0476; found 257.0481.

(2-Bromophenyl)(imidazo[1,2-a]pyridin-3-yl)methanone (**3am**).[14] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 150 mg, 50%); mp 165−166 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.79 (d, J = 6.8 Hz, 1H), 7.89 (s, 1H), 7.82 (d, J = 9.2 Hz, 1H), 7.70 (d, J = 7.6 Hz, 1H), 7.60 (t, J = 6.8 Hz, 1H), 7.50−7.43 (m, 2H), 7.39 (t, J = 7.6 Hz, 1H), 7.21 (t, J = 6.8 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 183.8, 149.6, 146.9, 140.4, 133.6, 131.4, 130.0, 129.2, 129.1, 127.2, 123.6, 119.9, 117.9, 115.6. ESI-MS: m/z [M + H]$^+$ 301.

Imidazo[1,2-a]pyridin-3-yl(naphthalen-1-yl)methanone (**3an**).[19a] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 196 mg, 72%); mp 133−134 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.92 (d, J = 5.6 Hz, 1H), 8.21 (d, J = 7.6 Hz, 1H), 8.03 (d, J = 8.4 Hz, 1H), 7.80 (s, 1H), 7.75 (d, J = 7.2 Hz, 1H), 7.73 (d, J = 8.0 Hz, 1H), 7.87 (dt, J = 8.8 Hz, 1H), 7.63−7.51 (m, 4H), 7.23 (t, J = 7.2 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 186.1, 149.3, 146.4, 136.7, 133.9, 131.3, 130.7, 129.9, 129.1, 128.5, 127.3, 127.1, 126.6, 125.3, 125.0, 124.5, 117.8, 115.5. ESI-MS: m/z [M + H]$^+$ 273.

Imidazo[1,2-a]pyridin-3-yl(naphthalen-2-yl)methanone (**3ao**).[17] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 201 mg, 74%); mp 127−128 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.81 (d, J = 6.8 Hz, 1H), 8.40 (s, 1H), 8.31 (s, 1H), 8.01−7.93 (m, 4H), 7.86 (d, J = 8.8 Hz, 1H), 7.65−7.53 (m, 3H), 7.18 (m, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184.8, 149.1, 145.6, 136.5, 135.1, 132.5, 130.0, 129.5, 129.3, 129.0, 128.7, 128.1, 127.9, 127.0, 125.1, 123.8, 117.8, 115.2. ESI-MS: m/z [M + H]$^+$ 273.

Furan-2-yl(imidazo[1,2-a]pyridin-3-yl)methanone (**3ap**). Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 91 mg, 43%); mp 149−150 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.81 (d, J = 6.8 Hz, 1H), 8.87 (s, 1H), 7.81 (d, J = 8.8 Hz, 1H), 7.71 (d, J = 0.8 Hz, 1H), 7.54 (t, J = 7.6 Hz, 1H), 7.40 (d, J = 3.2 Hz, 1H), 7.13 (t, J = 7.2 Hz, 1H), 7.63 (t, J = 2.0 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 170.5, 153. 0, 148.8, 145.9, 144.7, 129.4, 129.0, 122.5, 117.8, 117.5, 115.1, 112.4. HRMS (ESI): calcd for $C_{12}H_9N_2O_2$ m/z [M + H]$^+$ 213.0659; found 213.0657.

Imidazo[1,2-a]pyridin-3-yl(thiophen-2-yl)methanone (**3aq**).[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 100 mg, 44%); mp 111−112 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.65 (d, J = 7.2 Hz, 1H), 8.51 (s, 1H), 7.89 (d, J = 3.6 Hz, 1H), 7.81 (d, J = 9.2 Hz, 1H), 7.71 (d, J = 4.8 Hz, 1H), 7.54 (t, J = 7.2 Hz, 1H), 7.23 (t, J = 3.2 Hz, 1H), 7.13 (t, J = 7.2 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 175.7, 149.1, 143.8, 143.7, 132.6, 131.9, 129.3, 128.8, 128.0, 123.3, 117.9, 115.1. ESI-MS: m/z [M + H]$^+$ 229.

1-(Imidazo[1,2-a]pyridin-3-yl)-2,2-dimethylpropan-1-one (**3aw**).[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 125 mg, 62%); mp 137−138 °C; ^{1}H NMR (400 MHz, DMSO-d_6) δ 9.67 (d, J = 7.2 Hz, 1H), 8.73 (s, 1H), 7.83 (d, J = 9.2 Hz, 1H), 7.63 (t, J = 7.2 Hz, 1H), 7.26 (t, J = 7.2 Hz, 1H), 1.39 (s, 9H). ^{13}C{H} NMR (101 MHz, DMSO-d_6) δ 196.7, 147.6, 143.2, 129.7, 129.1, 121.4, 117.7, 115.8, 44.1, 28.8. ESI-MS: m/z [M + H]$^+$ 203.

(8-Methylimidazo[1,2-a]pyridin-3-yl)(phenyl)methanone (**3ba**).[14] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 172 mg, 73%); mp 85−86 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.61 (d, J = 7.2 Hz, 1H), 8.19 (s, 1H), 7.88 (d, J = 6.8 Hz, 2H), 7.62 (t, J = 7.6 Hz, 1H), 7.55 (t, J = 7.6 Hz, 2H), 7.35 (d, J = 6.8 Hz, 1H), 7.07 (t, J = 7.2 Hz, 1H), 2.71 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184.93, 149.3, 145.0, 139.4, 132.0, 130.9, 128.9, 128.6, 128.5, 127.6, 126.7, 115.2, 17.0. ESI-MS: m/z [M + H]$^+$ 237.

(8-Methoxyimidazo[1,2-a]pyridin-3-yl)(phenyl)methanone (**3ca**). Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 181 mg, 72%); mp 98−99 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.34 (d, J = 6.0 Hz, 1H), 8.15 (s, 1H), 7. 88 (d, J = 6.8 Hz, 2H), 7.62 (t, J = 4.8 Hz, 1H), 7.54 (t, J = 7.6 Hz, 2H), 7.05 (t, J = 7.6 Hz, 1H), 6.85 (d, J = 6.8 Hz, 1H), 4.08 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 185.1, 148.8, 144.2, 143.0, 139.3, 130.1, 128.9, 128.6, 124.5, 121.4, 115.2, 106.1, 56.2. HRMS (ESI): calcd for $C_{15}H_{13}N_2O_2$ m/z [M + H]$^+$ 253.0972; found 253.0976.

(7-Methylimidazo[1,2-a]pyridin-3-yl)(phenyl)methanone (**3ea**).[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 175 mg, 74%); mp 130−131 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.61 (d, J = 7.2 Hz, 1H), 8.15 (s, 1H), 7.86 (d, J = 7.6 Hz, 2H), 7.62−7.51 (m, 4H), 6.98 (d, J = 7.2 Hz, 1H), 2.51 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184.6, 149.6, 146.0, 141.2, 139.4, 131.9, 128.8, 128.6, 128.1, 123.3, 117.6, 116.4, 21.6. ESI-MS: m/z [M + H]$^+$ 237.

(7-Methoxyimidazo[1,2-a]pyridin-3-yl)(phenyl)methanone (**3fa**).[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 175 mg, 70%); mp 95−96 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.56 (d, J = 7.6 Hz, 1H), 8.11 (s, 1H), 7.86 (d, J = 6.8 Hz, 2H), 7.60 (t, J = 7.2 Hz, 1H), 7.53 (t, J = 4.0 Hz, 2H),7.12 (s, 1H), 6.83 (d, J = 7.6, 1.9 Hz, 1H), 3.95 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184.3, 161.3, 145.7, 139.3, 134.1, 131.9, 130.1, 129.5, 128.7, 123.1, 109.3, 95.6, 55.9. ESI-MS: m/z [M + H]$^+$ 253.

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*(6-Methylimidazo[1,2-a]pyridin-3-yl)(phenyl)methanone (**3ha**).*[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 175 mg, 74%); mp 95−96 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.58 (s, 1H), 8.16 (s, 1H), 7.87 (d, J = 7.6 Hz, 2H), 7.71 (d, J = 8.8 Hz, 1H), 7.61 (d, J = 7.2 Hz, 1H), 7.53 (t, J = 7.6 Hz, 2H), 7.40 (d, J = 8.0 Hz, 1H), 2.46 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184.8, 148.2, 145.6, 139.5, 132.4, 131.9, 128.8, 128.6, 126.9, 125.2, 123.4, 116.9, 18.4. ESI-MS: m/z [M + H]$^+$ 237.

*(6-Fluoroimidazo[1,2-a]pyridin-3-yl)(phenyl)methanone (**3ia**).*[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 173 mg, 72%); mp 120−121 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.76 (dd, J = 4.4, 2.4 Hz, 1H), 8.25 (s, 1H), 7.88 (d, J = 7.2 Hz, 2H), 7.81 (dd, J = 9.6, 4.8 Hz, 1H), 7.63 (t, J = 7.2 Hz, 1H), 7.55 (t, J = 7.2 Hz, 2H), 7.48 (td, J = 7.2, 2.4 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184.9, 154.6 (C−F, $^1J_{C-F}$ = 238 Hz), 146.5, 145.9 (C−F, $^4J_{C-F}$ = 2.4 Hz), 138.9, 132.3, 128.8, 128.7, 124.4, 120.9 (C−F, $^2J_{C-F}$ = 24.9 Hz), 118.0 (C−F, $^3J_{C-F}$ = 8.8 Hz), 116.2 (C−F, $^2J_{C-F}$ = 42.7 Hz). ESI-MS: m/z [M + H]$^+$ 241.

*(6-Chloroimidazo[1,2-a]pyridin-3-yl)(phenyl)methanone (**3ja**).*[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 174 mg, 68%); mp 129−130 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.85 (s, 1H), 8.22 (s, 1H), 7.88 (d, J = 6.8 Hz, 2H), 7.76 (d, J = 7.8 Hz, 1H), 7.63 (t, J = 7.2 Hz, 1H), 7.57−7.51 (m, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184.9, 145.6, 138.9, 132.3, 130.6, 128.8, 128.7, 126.9, 123.8, 123.6, 118.0. ESI-MS: m/z [M + H]$^+$ 257.

*(6-Iodoimidazo[1,2-a]pyridin-3-yl)(phenyl)methanone (**3ka**).*[14] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 212 mg, 61%); mp 146−147 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 10.04 (s, 1H), 8.14 (s, 1H), 7.87 (d, J = 7.2 Hz, 2H), 7.73 (d, J = 9.2 Hz, 1H), 7.65−7.52 (m, 4H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184.8, 147.6, 145.2, 138.9, 137.4, 133.8, 132.3, 128.8, 128.7, 123.3, 118.7, 78.6. ESI-MS: m/z [M + H]$^+$ 349.

*Phenyl(6-(trifluoromethyl)imidazo[1,2-a]pyridin-3-yl)-methanone (**3la**).*[16] Petroleum ether/ethyl acetate = 1:1; pale yellow powder (yield: 165 mg, 57%); mp 147−148 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 10.15 (s, 1H), 8.32 (s, 1H), 7.95 (d, J = 9.2 Hz, 2H), 7.90 (d, J = 7.2 Hz, 2H), 7.71 (dd, J = 1.6, 9.2 Hz, 1H), 7.66 (t, J = 7.2 Hz, 1H), 7.57 (t, J = 7.2 Hz, 2H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 185.1, 145.8, 138.5, 132.7, 128.9, 128.7127.9 (C−F, $^2J_{C-F}$ = 5.4 Hz), 125.5 (C−F, $^3J_{C-F}$ = 2.3 Hz), 124.2, 123.2 (C−F, $^1J_{C-F}$ = 270 Hz) 119.8, 119.5, 118.4. ESI-MS: m/z [M + H]$^+$ 291.

*(5-Methylimidazo[1,2-a]pyridin-3-yl)(phenyl)methanone (**3ma**).*[17] Petroleum ether/ethyl acetate = 1:1; light yellow oil (yield: 158 mg, 67%); ^{1}H NMR (400 MHz, chloroform-d) δ 8.06−8.03 (m, 3H), 7.76 (d, J = 8.8 Hz, 1H), 7.65 (t, J = 7.2 Hz, 1H), 7.56−7.50 (m, 3H), 6.95 (d, J = 6.8 Hz, 1H), 2.69 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 182.4, 150.7, 145.9, 140.1, 138.4, 133.0, 130.2, 129.6, 128.6, 125.8, 116.2, 115.2, 22.5. ESI-MS: m/z [M + H]$^+$ 237.

*Imidazo[1,2-a]quinolin-1-yl(phenyl)methanone (**3na**).*[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 158 mg, 58%); mp 160−161 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 8.44 (d, J = 8.4 Hz, 1H), 8.08 (d, J = 7.6 Hz, 2H), 7.99 (s, 1H), 7.84 (dd, J = 15.2, 8.0 Hz, 2H), 7.69−7.63 (m, 3H), 7.58−7.51 (m, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 184.3, 149.5, 146.5, 138.7, 133.5, 133.1, 131.4, 130.1, 129.1, 129.0, 128.6, 128.4, 125.8, 124.7, 119.9, 116.8. ESI-MS: m/z [M + H]$^+$ 273.

*Imidazo[2,1-a]isoquinolin-3-yl(phenyl)methanone (**3oa**).* Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 150 mg, 55%); mp 168−169 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.41 (d, J = 7.2 Hz, 1H), 8.76 (d, J = 6.0 Hz, 1H), 8.16 (s, 1H), 7.93 (d, J = 6.8 Hz, 2H), 7.84 (d, J = 6.0 Hz, 1H), 7.75−7.72 (m, 2H), 7.64 (t, J = 7.2 Hz, 1H), 7.56 (t, J = 7.6 Hz, 2H), 7.56 (d, J = 7.6 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 185.4, 147.2, 143.7, 139.3, 132.3, 131.1, 130.0, 129.0, 128.6, 128.6, 126.9, 125.2, 124.6, 124.5, 122.9, 115.1. HRMS (ESI): calcd for $C_{18}H_{13}N_2O$ m/z [M + H]$^+$ 273.1022; found 273.1025.

*Imidazo[1,2-b]isoquinolin-3-yl(phenyl)methanone (**3pa**).* Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 139 mg, 51%); mp 140−141 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.52 (d, J = 0.8 Hz, 1H), 9.04 (d, J = 4.5 Hz, 1H), 8.07 (dd, J = 7.9, 1.4 Hz, 1H), 7.62 (ddd, J = 8.0, 6.9, 1.0 Hz, 1H), 7.59−7.52 (m, 4H), 7.45−7.37 (m, 4H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 157.6, 155.1, 149.9, 149.7, 141.5, 132.8, 130.2, 129.4, 129.0, 128.6, 128.3, 127.7, 127.51, 127.0, 124.9, 117.5. HRMS (ESI): calcd for $C_{18}H_{13}N_2O$ m/z [M + H]$^+$ 273.1022; found 273.1028.

*Imidazo[1,2-b]pyridazin-3-yl(phenyl)methanone (**3qa**).*[16] Petroleum ether/ethyl acetate = 1:2; pale brown powder (yield: 112 mg, 50%); mp 157−158 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 8.68 (d, J = 3.2 Hz, 1H), 8.22 (s, 1H), 8.15 (d, J = 7.6 Hz, 1H), 7.95 (d, J = 7.2 Hz, 2H), 7.64 (t, J = 7.2 Hz, 1H), 7.54 (t, J = 7.6 Hz, 2H), 7.35 (dd, J = 9.2, 3.6 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 183.5, 144.6, 143.2, 142.7, 138.7, 132.7, 129.4, 128.6, 126.8, 126.2, 120.3. ESI-MS: m/z [M + H]$^+$ 224.

*Imidazo[1,2-a]pyrimidin-3-yl(phenyl)methanone (**3ra**).*[16] Petroleum ether/ethyl acetate = 1:2; pale brown powder (yield: 91 mg, 41%); mp 149−150 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 10.00 (d, J = 7.2 Hz, 1H), 8.84 (d, J = 2.4 Hz, 1H), 8.42 (s, 1H), 7.90 (d, J = 5.2 Hz, 2H), 7.65 (t, J = 7.2 Hz, 1H), 7.56 (t, J = 7.6 Hz, 2H), 7.25 (d, J = 3.6 Hz, 1H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 185.2, 153.7, 151.5, 146.4, 138.3, 136.7, 132.6, 128.9, 128.8, 121.8, 111.3. ESI-MS: m/z [M + H]$^+$ 224.

*(7-Methoxyimidazo[1,2-a]pyridin-3-yl)(3,4,5-trimethoxyphenyl)-methanone (**3fb′**).*[10] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 209 mg, 61%); mp 192−193 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 9.51 (d, J = 7.6 Hz, 1H), 8.18 (s, 1H), 7.13−7.11 (m, 3H), 6.84 (dd, J = 7.6, 2.4 Hz, 1H), 3.96 (s, 3H), 3.95 (s, 3H), 3.94 (s, 6H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 183.2, 161.1, 153.2, 151.4, 145.9, 141.5, 134.5, 129.4, 123.0, 109.1, 106.3, 95.8, 61.0, 56.3, 55.9. ESI-MS: m/z [M + H]$^+$ 343.

*3-(Pyridin-2-ylamino)-1-(p-tolyl)propan-1-one (**5ab**).*[16] Petroleum ether/ethyl acetate = 1:1; pale brown powder (yield: 144 mg, 60%); mp 79−80 °C; ^{1}H NMR (400 MHz, chloroform-d) δ 8.09 (d, J = 5.2 Hz, 1H), 7.86 (d, J = 8.0 Hz, 2H), 7.36 (t, J = 8.0 Hz, 1H), 7.25 (d, J = 7.6 Hz, 2H), 6.54 (t, J = 7.0 Hz, 1H), 6.38 (d, J = 8.4 Hz, 1H), 4.95 (br s, 1H), 3.83−3.78 (q, J = 6.4 Hz, 2H), 3.29 (t, J = 5.6 Hz, 2H), 2.40 (s, 3H). ^{13}C{H} NMR (101 MHz, chloroform-d) δ 199.3, 158.4, 148.0, 144.1, 137.2, 134.4, 129.3, 128.2, 112.8, 108.0, 38.1, 36.7, 21.7. ESI-MS: m/z [M + H]$^+$ 241.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.joc.1c00023.

Optimization of reaction conditions and ^{1}H NMR and ^{13}C NMR spectra of compounds **3** and **5ab** (PDF)

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Notes

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