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A FACILE PREPARATION OF IMIDAZO[1,2-*a*]PYRIDIN-3-AMINE DERIVATIVES VIA A THREE-COMPONENT REACTION WITH β-CYCLODEXTRIN–SO_3H AS CATALYST

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Abstract – Because heterogeneous catalysts have attracted great interest in organic chemistry, this paper reports facile β-cyclodextrin–SO_3H–catalyzed cyclization to form imidazo[1,2-*a*]pyridin-3-amine derivatives via a three-component reaction. The main advantages of this strategy include short reaction time, practical simplicity, and high yield, and the catalyst can be separated easily by filtration and reused at least four times.

INTRODUCTION

Imidazo[1,2-*a*]pyridine derivatives are an important class of fused heterocyclic compounds because of their potential biological pharmacological activities.[1–4] They have also been found in many drugs such as zolpidem, alpidem, zolimidine, olprinone, saripidem, and necopidem.[5,6] Therefore, preparation of imidazo[1,2-*a*]pyridines has attracted increasing attention, and several effective strategies have been developed for the construction of imidazo[1,2-*a*]pyridines.[5–12] Condensation of 2-aminoazines, aldehydes, and isocyanides is one convenient technique for synthesizing imidazo[1,2-*a*]pyridines. In past decades, several catalysts, such as montmorillonite K10 (microwave),[13] ZrCl,[14] $ZnCl_2$,[15] $SnCl_2$,[16] $RuCl_3$,[17] $LaCl_3•7H_2O$,[18] $InCl_3$,[19] cellulose sulfuric acid,[20] *p*-toluenesulfonic acid,[21] γ-Fe_2O_3@–SiO_2–OSO_3H,[22] sulfide nanotubes,[23] and bromodimethylsulfonium bromide[24] have been developed for condensation of 2-aminoazines, aldehydes, and isocyanides. In addition, some catalyst-free and solvent-free protocols have been developed.[25] These methodologies give good results in many instances. However, some of the synthetic strategies have high-temperature requirements and suffer from low yields of products, long reaction times, high cost catalysts, tedious preparation of catalysts, difficult recovery of homogeneous catalysts and separation of products. Hence, the development of efficient, simple, easy work-up protocol

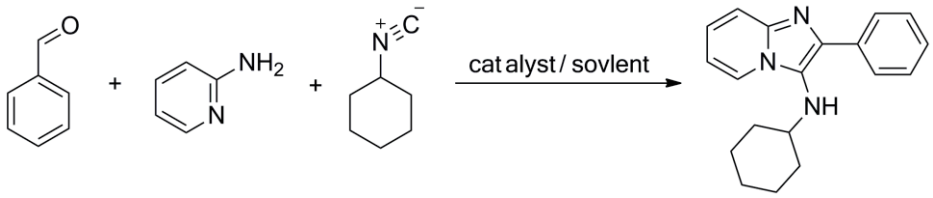
for the synthesis of imidazo[1,2-*a*]pyridine derivatives is still desirable and in demand.

β-Cyclodextrin (β-CD), a macrocyclic oligosaccharide with a hydrophobic cavity and a hydrophilic outer-surface, has attracted much attention and has been used widely.[26,27] Recently, β-CD and its derivatives have served as efficient catalysts in numerous organic reactions.[19,20,27–35] Because β-CD–SO_3H is a powerful catalyst, it is used for the synthesis of 4-thiazolidinones,[31] the esterification of carboxylic acids with alcohols,[36] and the preparation of 2,3-dihydroquinazolin-4(1*H*)-one derivatives in water,[34] and the catalytic property of β-CD–SO_3H has attracted our interests in organic synthesis.

Continuing our work on β-CD–SO_3H,[34] we present a cyclodextrin-SO_3H-catalyzed heterogeneous process to prepare imidazo[1,2-*a*]pyridin-3-amine derivatives via ring closure of substituted pyridin-2-amines, isocyanides, and aldehydes. To the best of our knowledge, this is a convenient means of preparing imidazo[1,2-*a*]pyridin-3-amine derivatives by using cheap, recyclable, and easily available β-CD–SO_3H as a heterogeneous catalyst.

RESULTS AND DISCUSSION

β-CD–SO_3H was prepared according to our previous work.[34] Subsequently, the effects of solvent, reaction time, and the amount of catalyst on the yields of products were investigated on the basis of the one-pot reaction of pyridin-2-amine with cyclohexanecarbonitrile and benzaldehyde (**Scheme 1**). The results are summarized in Table 1.



Scheme 1

Initially, the reaction was performed in aqueous media and catalyzed by β-CD–SO_3H (Table 1, Entry 1); unfortunately, this technique yielded only <35%. We then focused on screening organic solvents and noted the following: non-polar solvents, such as toluene, gave moderate yield of the product (Table 1, Entry 2); polar solvents gave much better yields than that of toluene (Table 1, Entries 3–7); both acetonitrile (Table 1, Entry 6) and ethanol (Table 1, Entry 7) gave excellent yields; and ethanol gave a slightly higher yield than that of acetonitrile. Reactions were conducted for determining the most-suitable temperature, and the results showed that, at room temperature to 60 °C, moderate yields (56–80%) could be obtained (Table 1, Entries 8–10), whereas the yield did not increase after reaction for 5 hours (Table 1, Entry 10). These results indicate that temperature is a key factor for this reaction. The reaction time was investigated further, and it was found that the yields were not obviously impacted when the reaction time

was changed from 2 hours to 1 hour (Table 1, Entries 7 and 11); however, the yield decreased substantially by shortening the reaction time to 30 min (Table 1, Entry 12). We next examined the effects of the amount of catalyst on this reaction, and the results (Table 1, Entries 13–15) indicated that this played an essential role in this reaction. Table 1, Entry 13, showed that the yield of the reaction was not increased by extending reaction time under catalyst-free conditions but that the yields could be improved by adding 5% of β-CD–SO_3H (Table 1, Entry 14). By adding >10% of the catalyst, reaction occurred smoothly and resulted in a higher yield (up to 93%) (Table 1, Entry 11); however, an increase in the amount of catalyst cannot enhance the yield of the product (Table 1, Entry 15). Moreover, β-CD (Table 1, Entry 16) was also used as catalyst, a hydrogen bonding formed between "-CH_2OH" of β-CD and "O" at aldehyde, this bonding can enhances the eletrophilicity of carbonyl and accelerate proceeding of the reaction,[37,38] but the yield was much lower than that of β-CD–SO_3H. After completion of this reaction, β-CD–SO_3H could be isolated completely by filtration, and the organic phase was distilled in vacuum to yield the product, which was washed with 2 mL of ethyl acetate and cyclohexane (1:3 in volume).

Table 1. Optimization for synthesis of imidazo[1,2-*a*]pyridin-3-amine derivtives (model reaction) [a]

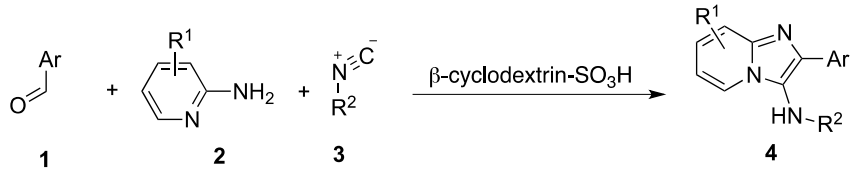
Entry	Solvent	Time (h)	Temperature (°C)	Amount of the catalyst (mol%)	Yields (%) [b]
1	H_2O	2	80	10 [c]	<35
2	toluene	2	80	10 [c]	65
3	CH_2Cl_2	2	reflux	10 [c]	45
4	THF	2	reflux	10 [c]	70
5	MeOH	2	reflux	10 [c]	75
6	MeCN	2	80	10 [c]	90
7	EtOH	2	reflux	10 [c]	94
8	EtOH	2	60	10 [c]	80
9	EtOH	2	40	10 [c]	74
10	EtOH	5	rt	10 [c]	56
11	EtOH	1	reflux	10 [c]	93
12	EtOH	0.5	reflux	10 [c]	84
13	EtOH	4	reflux	— [d]	35
14	EtOH	1	reflux	5 [c]	75
15	EtOH	1	reflux	20 [c]	94
16	EtOH	1	reflux	10 [e]	70

[a] All reactions were conducted with 2-aminobenzamide (1 mmol), benzaldehyde (1 mmol), and isocyanides (1 mmol); [b] Isolated yields; [c] β-cyclodextrin–SO_3H; [d] Catalyst-free; [e] β-cyclodextrin.

With these optimized conditions accessible, the protocol was applied further to various substituted pyridin-2-amine, isocyanides, and aldehydes. Generally, the reactions were performed by using 10 mol% of β-CD–SO_3H in ethanol or acetonitrile under refluxing conditions for 1 hour to achieve the desired yield, the results are listed in Table 2. It was observed that most of the reactions proceeded smoothly. First,

different aromatic aldehydes were used to react with pyridin-2-amine and isocyanocyclohexane, which afforded excellent yields of the products **4b** to **4h** (89–95%). Then, 2-isocyano-2-methylpropane was used instead of isocyanocyclohexane (Table 2, compounds **4i** to **4q**), and the protocols were also performed smoothly. The substituents on pyridin-2-amine (Table 2, compounds **4m** to **4r**) showed basically no influence on the yields of products. Moreover, the reactions of the ethyl 2-isocyanoacetate with pyridin-2-amine and aldehydes also gave excellent yields.

Table 2. β-Cyclodextrin–SO_3H catalyzed synthesis of imidazo[1,2-*a*]pyridin-3-amine derivatives [a]



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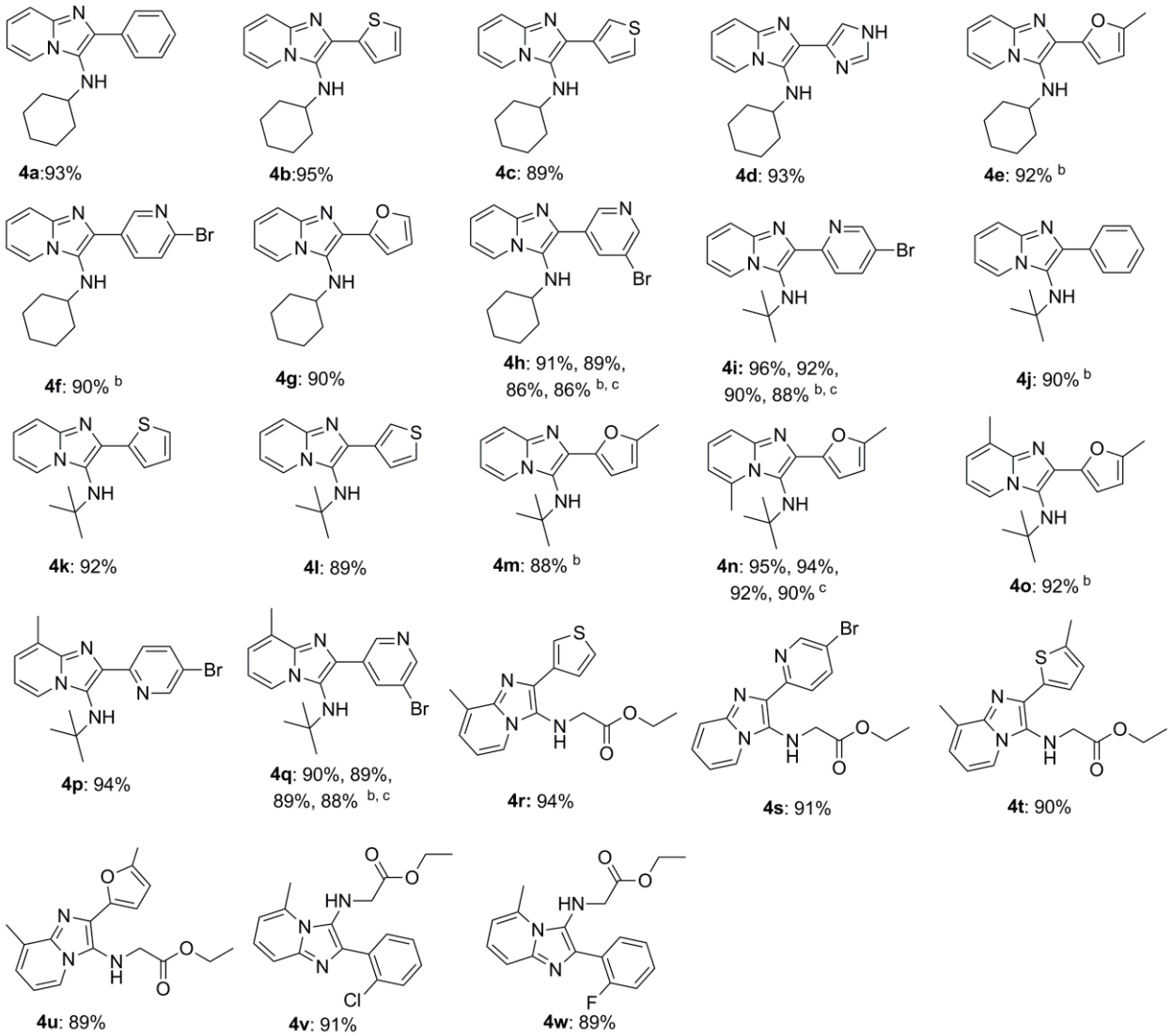


[a] Reaction conditions: substituted pyridin-2-amine (1 mmol), arylaldehyde (1 mmol), isocyanide (1 mmol), and β-cyclodextrin–SO_3H (mmol 10%); [b] Reactions performed in acetonitrile; [c] Same catalyst used for each of the four runs.

Subsequently, we turned our attention to catalyst recycling and reuse. After completing the reaction, the reaction mixture turned clear, and the catalysts were deposited on the button, which had been recovered by filtration, washed with CH_2Cl_2 and were reused in the next reaction. These results in Table 2 (compounds **4h**, **4i**, **4n**, and **4q**) indicated that the catalytic activity were maintained, even when the catalyst was reused 4 times, with a slight decrease in product yields.

To compare the efficiency of β-CD–SO_3H with the reported catalysts for the synthesis of imidazo[1,2-*a*]pyridin-3-amine derivatives, we tabulated the results of these catalysts to promote the synthesis of target compounds from substituted pyridin-2-amines, isocyanides, and aldehydes in Table 3. The results showed that β-CD–SO_3H is a better catalyst with respect to reaction. From the results of sulfonated catalyst (Table 3, Entries 5–7), it can be concluded that cellulose sulfuric acid,[20] sulfide nanotubes,[23] and β-CD–SO_3H could give excellent yields of the products, but the biggest differences in course of reaction are the reaction time and temperature, it concluded that β-CD–SO_3H could give much shorter reaction time than that of cellulose sulfuric acid; although the sulfide nanotubes could provide a little superiority in reaction time, but the yields of products were certain decreased when compared that of β-CD–SO_3H; moreover, the preparation process of β-CD–SO_3H[34] is much convenient than that of sulfide nanotubes.[23]

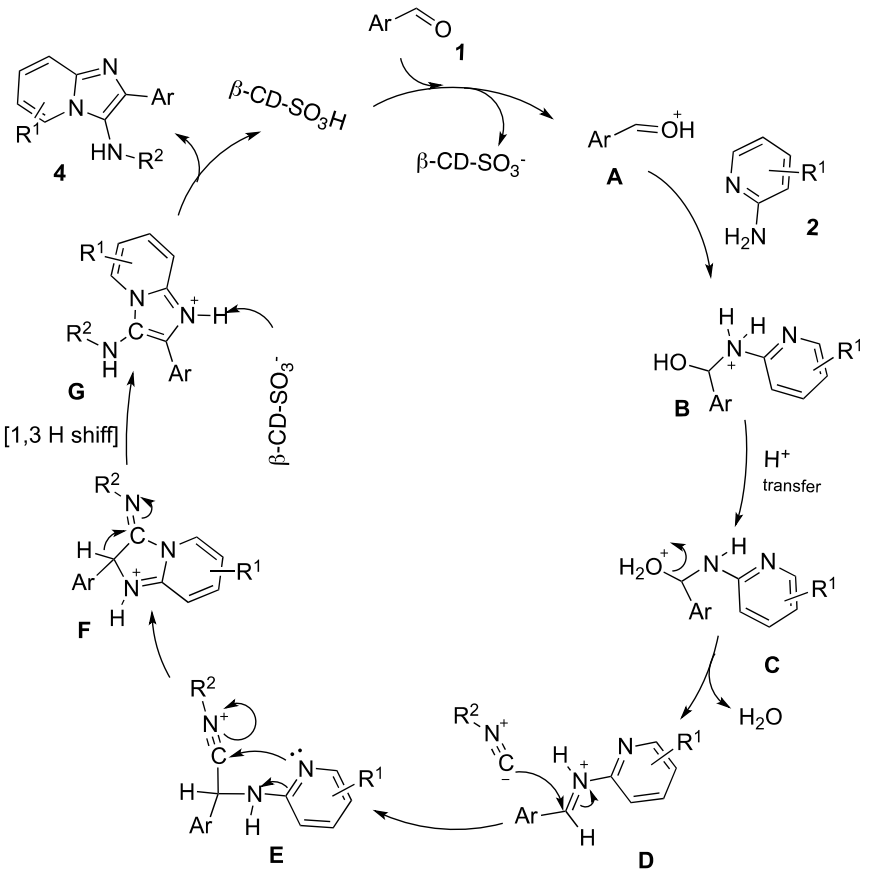
Table 3. Comparison of β-cyclodextrin–SO_3H with the reported catalysts for synthesis of imidazo[1,2-*a*]pyridin-3-amines

Entry	Catalyst	Solvent	Temperature (°C)	Time	Yield (%)	Reference
1	$ZrCl_4$	PEG-400	50	2–7 h	72−90	14
2	$ZnCl_2$	1,4-dioxane	reflux	1 h [a], 5 h [b]	9–75	15
3	$RuCl_3$	— [c]	40	1 h	87−93	17
4	$LaCl_3{\cdot}7H_2O$	— [c]	60	20–90 min	73−96	18
5	cellulose sulfuric acid	MeOH	rt [d]	3 h	89−98	20
6	sulfide nanotubes	MeOH	rt [d]	40 min	71−91	23
7	β-cyclodextrin–SO_3H	EtOH/ MeCN	reflux	1 h	89−96	This work

[a] Microwave; [b] Reflux; [c] Solvent-free; [d] Room temperature.

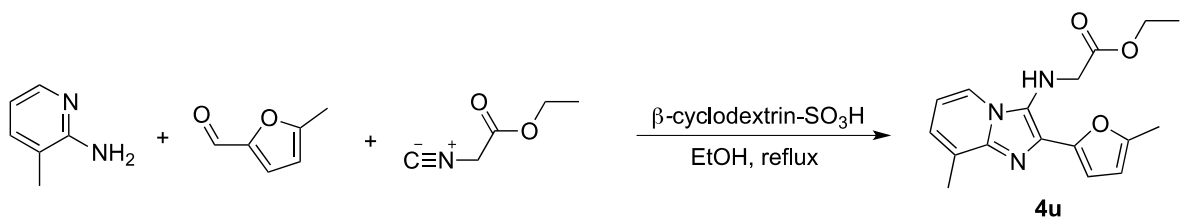
Some possible mechanisms for construction of imidazo[1,2-*a*]pyridin-3-amine derivatives via three-component one-pot reaction have been reported previously.[18,39] According to our experimental observations and also other mechanisms reported in the literature,[39] a possible mechanism of this reaction is presented in **Scheme 2**. First, the aldehyde (1) was activated by β-CD–SO_3H to give a positive ion, **A**, and β-CD–SO_3^-. Then, the carbonyl unit of **A** was attacked by *N*-nucleophilic amine (**2**) to produce an intermediate, **B**, which in turn gave an intermediate, **C**. After which, intermediate **D** was formed via dewatering of intermediate **C**, which was attacked further by a carbonyl unit of isocyanide to generate

intermediate **E**. Finally, intermediate **G** was formed via rearrangement and proton transfer, which in turn affords the target product **4**. The generated β-CD–SO_3^- subsequently became β-CD–SO_3H by receiving a proton, which could then be used in the next catalytic cycle.



Scheme 2

To check the applicability of this protocol, a gram scale-up reaction for the synthesis of ethyl 2-((8-methyl-2-(5-methylfuran-2-yl)imidazo[1,2-*a*]pyridin-3-yl)amino)acetate was performed (**Scheme 3**), which resulted in a 90% yield (2.65 g). Because both carboxylic acid and acylhydrazine can be derived from compounds containing an ester and can form further by obtaining molecules such as amide and hydrazone to develop biologically active molecules, our continuing research focuses on deriving and screening for biological activity based on these ester-containing compounds.



Scheme 3

EXPERIMENTAL

The substituted pyridin-2-amines were obtained from TCI (Shanghai, China), isocyanides aromatic aldehydes were purchased from Accela ChemBio Co., Ltd (Shanghai, China). Melting points were uncorrected and determined on a WRX-4 monocular microscope (Shanghai Yice Apparatus & Equipment Co., Ltd, China). The ^{1}H-NMR and ^{13}C-NMR spectra were recorded on a JEOL ECX 500 NMR spectrometer (JEOL Ltd, Japan) at room temperature operating at 500 MHz for ^{1}H-NMR and 125 MHz for ^{13}C-NMR by using $CDCl_3$, DMSO-d_6 or CD_3OD as solvents and TMS as an internal standard; infrared spectra were recorded in KBr on a IR Pristige-21 spectrometer (Shimadzu corporation, Japan), absorbencies are reported in cm^{-1}; HR-MS were recorded on a Orbitrap LC-MS instrument (Q-Exative, Thermo Scientific™, American). The course of the reactions was monitored by TLC; analytical TLC was performed on silica gel GF 254.

Preparation of sulfonated β-cyclodextrin. To a well stirred mixture of β-cyclodextrin (10.0 g, 4.5 mmol) in CH_2Cl_2 (50 mL), chlorosulfonic acid (2.00 g, 10 mmol) was added slowly at 0 °C during 3 h. The resulting mixture was stirred for another 2 h to remove HCl from the reaction vessel. Then, the mixture was filtered and washed with MeOH and dried at room temperature to obtain β-cyclodextrin-SO_3H as a white powder. ^{1}H NMR (500 MHz, DMSO-d_6) δ 5.70 (d, J = 6.9 Hz, 7H), 5.65 (d, J = 2.4 Hz, 7H), 4.79 (d, J = 3.6 Hz, 7H), 4.43 (t, J = 5.7 Hz, 7H), 3.74-3.47 (m, 28H), 3.35-3.31 (m, 7H), 3.28-3.22 (m, 7H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 102.48, 82.06, 73.57, 72.94, 72.57, 60.43. The –SO_3H content was measured by titration method and showed 0.52 mequiv./g.[34]

General procedure for the preparation of imidazo[1,2-*a*]pyridin-3-amines. To a mixture of 2-aminopyridines (1 mmol), aromatic aldehydes (1 mmol) and isocyanides in EtOH (or MeCN) was added β-cyclodextrin-SO_3H (10 mol%). The reaction mixture was then allowed to stir for 1 h under 80 °C. After complication of this reaction, the resulting mixture was cooled and the β-cyclodextrin-SO_3H was removed by filtration, the organic phase was concentrated under reduced pressure. Afterwards the residue were washed with EtOAc and cyclohexane (1:3) and dried to give the product.

***N*-Cyclohexyl-2-phenylimidazo[1,2-*a*]pyridin-3-amine (4a):** White solid; mp 199.2-201.3 °C, lit.[20] 200-202 °C; IR 3257, 2957, 2925, 2852, 1630, 1554, 1503, 1447, 1342, 1226, 1197, 1099, 1017, 889, 840, 752, 734, 605 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$) δ 1.12-1.25 (m, 5H), 1.68-1.83 (m, 5H), 2.95-3.0 (m, 1H), 3.12 (br, 1H), 6.76 (t, J = 5 Hz, 1H), 7.2 (t, J = 5 Hz, 1H), 7.32 (t, J = 10 Hz, 1H), 7.43-7.46 (m, 2H), 7.54 (d, J = 5 Hz, 1H), 8.02-8.04 (m, 2H), 8.10 (d, J = 5 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 24.9, 25.8, 34.3, 57.0, 111.6, 117.3, 122.8, 123.9, 127.1, 127.3, 128.6, 134.6, 141.7, 141.8; HR-MS (ESI^{+}) *m/z* Calcd for $C_{19}H_{21}N_3$ $[M+H]^+$ 292.18082; Found 292.18054.

***N*-Cyclohexyl-2-(thiophen-2-yl)imidazo[1,2-*a*]pyridin-3-amine (4b):** Grey white solid; mp 164.3-165.4 °C; IR 3305, 2967, 2927, 2854, 1648, 1556, 1502, 1344, 1226, 1197, 1099, 1019, 901, 845,

753, 737, 602 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$) δ 1.12-1.36 (m, 6H), 1.60-1.88 (m, 4H), 3.04-3.08 (m, 1H), 3.10 (br, 1H, NH), 6.78 (t, *J*= 10 Hz, 1H), 7.12-7.17 (m, 2H), 7.32 (d, *J* = 4.5 Hz, 1H), 7.54 (d, *J* = 10 Hz, 1H), 7.62 (d, *J* = 5 Hz, 1H), 8.10 (d, *J* = 5 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 25.0, 25.8, 34.4, 57.2, 112.1, 116.9, 122.9, 124.1, 124.3, 124.8, 124.9, 125.0, 127.8, 141.5; HR-MS (ESI^+) *m/z* Calcd for $C_{17}H_{19}N_3S$ $[M+H]^+$ 298.13724; Found 298.13699.

***N*-Cyclohexyl-2-(thiophen-3-yl)imidazo[1,2-*a*]pyridin-3-amine (4c):** Yellow solid; mp 170.8-171.6 °C; IR 3309, 3301, 2955, 2863, 1632, 1549, 1458, 1342, 1226, 1197, 1099, 1017, 889, 840, 752, 734, 605 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$) δ 1.16-1.37 (m, 5H), 1.70-1.85 (m, 5H), 2.97-3.02 (m, 1H), 3.09 (br, 1H), 6.77 (t, *J* = 6.5 Hz, 1H), 7.12 (t, *J* = 7.5 Hz, 1H), 7.38 (dd, *J* = 3 Hz, 5 Hz, 1H), 7.54 (d, *J* = 9 Hz, 1H), 7.84 (dd, *J* = 3 Hz, 1 Hz, 1H), 8.09 (d, *J* = 7 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 25.0, 25.8, 34.4, 57.2, 112.0, 117.0, 120.0, 121.9, 122.9, 124.3, 124.6, 125.8, 126.6; HR-MS (ESI^+) *m/z* Calcd for $C_{17}H_{19}N_3S$ $[M+H]^+$ 298.13724; Found 298.13687.

***N*-Cyclohexyl-2-(1*H*-imidazol-4-yl)imidazo[1,2-*a*]pyridin-3-amine (4d):** Yellow solid; mp 184.2-185.7 °C; IR 3286, 2959, 2932, 2857, 1633, 1559, 1513, 1467, 889, 843, 756, 714, 625 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$) δ 1.13-1.34 (m, 5H), 1.70-1.86 (m, 5H), 2.96-3.00 (m, 1H), 4.19 (br, 1H), 6.85 (t, *J* = 10 Hz, 1H), 7.20 (t, *J* = 5 Hz, 1H), 7.61 (d, *J* = 10 Hz, 1H), 7.65 (s, 1H), 7.74 (s, 1H), 8.08 (d, *J* = 5 Hz, 1H), 9.81 (br, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 25.09, 25.77, 34.14, 56.66, 112.82, 115.89, 118.53, 123.04, 125.51, 125.54, 125.82, 127.05, 130.70, 135.51, 139.31, 140.30; HR-MS (ESI^+) *m/z* Calcd for $C_{16}H_{19}N_5$ $[M+H]^+$ 282.17132; Found 282.17108.

***N*-Cyclohexyl-2-(5-methylfuran-2-yl)imidazo[1,2-*a*]pyridin-3-amine (4e):** White solid; mp 89.1-90.5 °C; IR 3305, 2968, 2945, 2856, 1645, 1545, 1543, 1450, 1354, 1226, 1098, 1013, 885, 837, 755 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$): 1.14-1.32 (m, 6H), 1.71-1.89 (m, 4H), 2.39 (s, 1H), 2.91-2.98 (m, 1H), 3.50 (br, 1H), 6.09-6.10 (m, 1H), 6.72 (d, *J* = 3.1 Hz, 1H), 6.75 (td, *J* = 6.7, 0.8 Hz, 1H), 7.09 (ddd, *J* = 9.0, 6.7, 1.0 Hz, 1H), 7.49 (d, *J* = 9.0 Hz, 1H), 8.05 (dd, *J* = 6.8, 1.1 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 13.9, 25.1, 25.9, 34.3, 57.3, 107.4, 107.6, 111.6, 117.2, 122.8, 123.9, 125.0, 141.9, 148.5, 151.5; HR-MS (ESI^+) *m/z* Calcd for $C_{18}H_{21}N_3O$ $[M+H]^+$ 296.17574; Found 296.17548.

2-(6-Bromopyridin-3-yl)-*N*-cyclohexylimidazo[1,2-*a*]pyridin-3-amine (4f): Yellow solid; mp 156.4-158.3 °C; IR 3287, 3007, 2946, 2872, 1645, 1564, 1521, 1453, 1374, 1246, 1213, 1106, 1034, 886, 844, 755, 745, 632 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$) δ 1.13-1.33 (m, 6H), 1.67-1.81 (m, 4H), 2.92-2.97 (m, 1H), 3.15 (br, *J* = 5 Hz, 1H), 6.82 (t, *J* = 5 Hz), 7.18 (t, *J* = 5 Hz), 8.06 (dd, *J* = 7 Hz, 1 Hz, 1H), 8.31 (d, *J* = 2.5 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 24.9, 25.7, 34.3, 57.0, 112.4, 117.6, 120.00, 120.01, 122.9, 125.1, 125.5, 128.1, 129.9, 133.0, 137.0, 140.5, 142.2, 148.3; HR-MS (ESI^+) *m/z* Calcd for $C_{18}H_{19}BrN_4$ $[M+H]^+$ 371.08659; Found 371.08649.

***N*-Cyclohexyl-2-(furan-2-yl)imidazo[1,2-*a*]pyridin-3-amine (4g):** Yellow solid; mp 121.3-123.2 °C,

lit.[14] 121-122 °C; IR 3308, 2978, 2944, 2867 1645, 1538, 1533, 1467, 1354, 1226, 1209, 1109, 1065, 878, 843, 759, 738, 635 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$) δ 1.12-1.31 (m, 6H), 1.71-1.91 (m, 4H), 2.91-2.97 (m, 1H), 3.61 (d, *J* = 6.7 Hz, 1H), 6.52 (dd, *J* = 3.4, 1.8 Hz, 1H), 6.76 (td, *J* = 6.8, 1.1 Hz, 1H), 6.86 (dd, *J* = 3.4, 0.7 Hz, 1H), 7.11 (ddd, *J* = 9.1, 6.6, 1.3 Hz, 1H), 7.49 (ddt, *J* = 3.7, 1.9, 1.0 Hz, 2H), 8.04 (dt, *J* = 6.9, 1.2 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 25.1, 25.8, 34.2, 57.2, 106.5, 111.6, 111.8, 117.3, 122.9, 124.1, 125.6, 128.0, 141.6, 141.9, 150.4; HR-MS (ESI^+) *m/z* Calcd for $C_{17}H_{19}N_3O$ [M+H]+ 282.16009; Found 282.16000.

2-(5-Bromopyridin-3-yl)-*N*-cyclohexylimidazo[1,2-*a*]pyridin-3-amine (4h): Yellow solid; mp 158.8-159.6 °C; IR 3273, 2987, 2955, 2892, 1639, 1564, 1576, 1457, 1335, 1246, 1209, 1089, 1027, 892, 844, 752, 734 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$) δ 1.12-1.31 (m, 6H), 1.68-1.82 (m, 4H), 2.92-2.98 (m, 1H), 3.07 (d, *J* = 4.5 Hz, 1H), 6.82 (td, *J* = 6.8, *J* = 1.0 Hz, 1H), 7.18 (ddd, *J* = 9.0, 6.7, 1.3 Hz, 1H), 7.53 (dd, *J* = 9.1, 1.0 Hz, 1H), 8.07 (dd, *J* = 6.9, 1.1 Hz, 1H), 8.63 (t, *J* = 2.1 Hz, 1H), 8.63 (t, *J* = 2.1 Hz, 1H), 9.26 (d, *J* = 1.8 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 24.8, 25.7, 34.3, 57.1, 112.4, 112.7, 117.7, 121.7, 122.9, 125.0, 125.9, 132.3, 132.6, 136.8, 142.3, 146.0, 149.0; HR-MS (ESI^+) *m/z* Calcd for $C_{18}H_{19}BrN_4$ $[M+H]^+$ 371.08659; Found 371.08646.

2-(5-Bromopyridin-2-yl)-*N*-(*tert*-butyl)imidazo[1,2-*a*]pyridin-3-amine (4i): Yellow solid; mp 124.1-124.9 °C; IR 3299, 2987, 2945, 2878, 1656, 1564, 1523, 1452, 1352, 1233, 1207, 1102, 1027, 894, 854, 755, 739, 645 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$) δ 1.14 (s, 9H), 5.18 (br, 1H), 6.75 (t, *J* = 5 Hz, 1H), 7.18 (t, *J* = 6.5 Hz, 1H), 8.06 (dd, *J* = 9 Hz, 6.5 Hz, 1H), 7.52 (d, *J* = 9 Hz, 1H), 8.09 (d, *J* = 8.5 Hz, 1H), 8.27 (d, *J* = 7 Hz, 1H), 8.27 (d, *J* = 2 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 30.2, 57.4, 111.8, 117.3, 118.5, 122.7, 124.4, 124.9, 129.1, 133.4, 139.3, 141.6, 149.4, 153.1; HR-MS (ESI^+) *m/z* Calcd for $C_{16}H_{17}BrN_4$ $[M+H]^+$ 345.07094; Found 345.07098.

***N*-(*tert*-Butyl)-2-phenylimidazo[1,2-*a*]pyridin-3-amine (4j):** White solid; mp 166.2-167.9 °C, lit.[38] 169-170 °C; IR 3320, 2989, 2965, 2862, 1645, 1554, 1523, 1468, 1342, 1226, 1197, 1099, 1017, 840, 752, 734 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$) δ 1.02 (s, 9H), 5.64 (br, 1H), 6.79 (dd, *J* = 9.3, 3.9 Hz, 1H), 7.33-7.28 (m, 1H), 7.40-7.43 (m, 2H), 7.58 (d, *J* = 8.6 Hz, 1H), 7.89 (s, 1H), 7.90 (s, 1H), 8.25 (d, *J* = 6.8 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 30.4, 56.6, 111.8, 117.0, 123.7, 124.8, 127.7, 128.3, 128.4, 134.6, 138.9, 141.7; HR-MS (ESI^+) *m/z* Calcd for $C_{17}H_{19}N_3$ $[M+H]^+$ 266.16517; Found 266.16495.

***N*-(*tert*-Butyl)-2-(thiophen-2-yl)imidazo[1,2-*a*]pyridin-3-amine (4k):** Yellow solid; mp 132.5-134.3 °C; IR 3305, 2968, 2945, 2856, 1645, 1564, 1523, 1457, 1340, 1236, 1230, 1109, 1031, 889, 832, 738 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$) δ 1.18 (s, 9H), 3.12 (br, 1H), 6.77 (td, *J* = 6.8, 1.0 Hz, 1H), 7.09 (dd, *J* = 5.0, 3.6 Hz, 1H), 7.14 (ddd, *J* = 9.0, 6.7, 1.2 Hz, 1H), 7.31 (dd, *J* = 5.1, 1.1 Hz, 1H), 7.53 (d, *J* = 9.0 Hz, 1H), 7.60 (dd, *J* = 3.6, 1.1 Hz, 1H), 8.21 (dt, *J* = 6.9, 1.1 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ ppm = 30.6, 56.9, 111.8, 113.1, 117.0, 123.0, 123.7, 124.9, 125.1, 127.5, 134.4, 137.1, 142.0;

HR-MS (ESI$^+$) *m/z* Calcd for $C_{15}H_{17}N_3S$ $[M+H]^+$ 272.12159; Found 272.12128.

***N*-(*tert*-Butyl)-2-(thiophen-3-yl)imidazo[1,2-*a*]pyridin-3-amine (4l):** White solid; mp 145.2-147.1 °C; IR 3308, 2970, 2947, 2878, 1655, 1556, 1543, 1467, 1342, 1236, 1233, 1116, 1041, 892, 834, 738 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$) δ 1.18 (s, 9H), 3.09 (s, 1H), 6.79 (t, *J* = 6.7 Hz, 1H) , 7.14 (dd, *J* = 8.3, 7.4 Hz, 1H), 7.39 (dd, *J* = 5.0, 3.0 Hz, 1H), 7.55 (d, *J* = 9.0 Hz, 1H), 7.74 (dd, *J* = 5.0, 1.2 Hz, 1H) 7.85 (dd, *J* = 2.9, 1.1 Hz, 1H), 8.10 (d, *J* = 6.8 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 34.4, 57.2, 112.0, 117.0, 121.9, 122.9, 124.3, 124.6, 125.8, 126.6, 133.3, 135.0, 141.4; HR-MS (ESI$^+$) *m/z* Calcd for $C_{15}H_{17}N_3S$ $[M+H]^+$ 272.12159; Found 272.12140.

***N*-(*tert*-Butyl)-2-(5-methylfuran-2-yl)imidazo[1,2-*a*]pyridin-3-amine (4m):** White solid; mp 114.3-115.4 °C; IR 3289, 2947, 2955, 2876, 1636, 1556, 1523, 1487, 1363, 1230, 1221, 1123, 894, 845, 756, 734 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$) δ 1.16 (s, 9H), 2.37 (s, 3H), 3.48 (br, 1H), 6.09 (m, 1H), 6.74 (td, *J* = 6.8, 1.0 Hz, 1H), 6.78 (d, *J* = 3.2 Hz, 1H), 7.12 (ddd, *J* = 8.9, 6.7, 1.2 Hz, 1H), 7.52-7.47 (m, 1H), 8.22 (dd, *J* = 6.0, 0.9 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 13.9, 30.2, 56.7, 107.6, 108.5, 111.5, 116.8, 123.6, 123.7, 124.7, 130.6, 142.1, 148.2, 151.5; HR-MS (ESI$^+$) *m/z* Calcd for $C_{16}H_{19}N_3O$ $[M+H]^+$ 270.16009; Found 270.15982.

***N*-(*tert*-Butyl)-5-methyl-2-(5-methylfuran-2-yl)imidazo[1,2-*a*]pyridin-3-amine (4n):** Brown solid; mp 72.3-73.2 °C; IR 3303, 2967, 2935, 2862, 1642, 1567, 1523, 1465, 1342, 1230, 889, 825, 752 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$) δ 1.14 (s, 9H), 2.19 (br, 1H), 2.38 (s, 3H), 2.60 (s, 3H), 6.08 (dd, *J* = 3.2, 1.0 Hz, 1H), 6.67 (t, *J* = 6.8 Hz, 1H), 6.79 (d, *J* = 3.2 Hz, 1H), 6.91 (dt, *J* = 6.75 Hz, 1.15 Hz, 1H), 8.13-8.08 (d, *J* = 6.85 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 13.85, 16.77, 30.18, 56.52, 107.50, 108.58, 111.44, 113.55, 121.65, 123.32, 124.18, 126.72, 142.55, 148.40, 151.30; HR-MS (ESI$^+$) *m/z* Calcd for $C_{17}H_{21}N_3O$ $[M+H]^+$ 284.17574; Found 284.17548.

***N*-(*tert*-Butyl)-8-methyl-2-(5-methylfuran-2-yl)imidazo[1,2-*a*]pyridin-3-amine (4o):** Brown sticky liquid; IR 3278, 2987, 2975, 2878, 2854, 1635, 1564, 1554, 1457, 1343, 1230, 1210, 845, 734 cm^{-1}; ^{1}H NMR (500 MHz, $DOCD_3$) δ 0.94 (s, 9H), 2.36 (s, 1H), 2.96 (s, 3H), 6.12 (dd, *J* = 3.2, 1.0 Hz, 1H), 6.62-6.57 (m, 1H), 6.69 (d, *J* = 3.2 Hz, 1H), 7.13 (dd, *J* = 9.0, 6.8 Hz, 1H), 7.25 (dd, *J* = 5.3, 4.3 Hz, 1H); ^{13}C NMR (125 MHz, $DOCD_3$) δ 12.25, 19.55, 28.25, 55.74, 107.17, 108.99, 111.89, 113.67, 114.34, 125.76, 131.82, 137.92, 143.11, 147.52, 151.72; HR-MS (ESI$^+$) *m/z* Calcd for $C_{17}H_{21}N_3O$ $[M+H]^+$ 284.17574; Found 284.17542.

2-(5-Bromopyridin-2-yl)-*N*-(*tert*-butyl)-8-methylimidazo[1,2-*a*]pyridin-3-amine (4p): Yellow solid; mp 101.5-103.4 °C; IR 3301, 2967, 2945, 2864, 2874, 1654, 1545, 1523, 1454, 1343, 1226, 1214, 1108, 895, 845, 757 cm^{-1}; ^{1}H NMR (500 MHz, $CDCl_3$) δ 1.13 (s, 9H), 2.61 (s, 3H), 5.16 (br, 1H), 6.66 (dt, *J* = 11.6, 5.8 Hz, 1H), 6.91 (dd, *J* = 6.6, 1.0 Hz, 1H), 7.87 (dt, *J* = 8.5, 2.7 Hz, 1H), 8.14 (d, *J* = 6.9 Hz, 1H), 8.18 (dd, *J* = 8.6, 3.5 Hz, 1H), 8.60 (d, *J* = 2.2 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 16.74, 30.17,

57.26, 118.25, 122.31, 123.01, 123.37, 127.21, 129.42, 139.17, 139.18, 142.12, 149.35, 153.56; HR-MS (ESI^+) *m/z* Calcd for $C_{17}H_{19}BrN_4$ $[M+H]^+$ 359.08659; Found 359.08624.

2-(4-Bromopyridin-3-yl)-*N*-(*tert*-butyl)-8-methylimidazo[1,2-*a*]pyridin-3-amine (4q): White solid; mp 163.1-164.5 °C; IR 3297, 2959, 2945, 2854, 1645, 1564, 1524, 1446, 1354, 1230, 1208, 1114, 902, 854, 755 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$) δ 1.06 (s, 9H), 2.61 (s, 3H), 6.73 (t, *J* = 6.8 Hz, 1H), 7.04-6.88 (m, 1H), 7.58-7.46 (m, 1H), 8.04 (d, *J* = 6.9 Hz, 1H), 8.25 (dd, *J* = 8.3, 2.4 Hz, 1H), 9.03 (dd, *J* = 2.5, 0.7 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 16.64, 30.59, 56.61, 112.08, 121.36, 123.72, 124.44, 127.42, 127.87, 130.99, 135.05, 138.11, 140.38, 143.03, 149.43; HR-MS (ESI^+) *m/z* Calcd for $C_{17}H_{19}BrN_4$ $[M+H]^+$ 359.08659; Found 359.08643.

Ethyl 2-((5-methyl-2-(thiophen-3-yl)imidazo[1,2-*a*]pyridin-3-yl)amino)acetate (4r): Brown sticky liquid; IR 3307, 2998, 2965, 2867, 1715, 1647, 1563, 1543, 1467, 1362, 1226, 1197, 1103, 1054, 895, 847, 752 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$) δ 1.25 (t, *J* = 7.1 Hz, 3H), 2.91 (s, 3H), 3.75 (d, *J* = 4.9 Hz, 2H), 3.80 (br, 1H), 4.21 (q, *J* = 7.2 Hz, 2H), 6.47 (dd, *J* = 6.8, 1.0 Hz, 1H), 7.05 (dd, *J* = 8.9, 6.9 Hz, 1H), 7.37-7.33 (m, 1H), 7.42 (d, *J* = 9.3 Hz, 1H), 7.71 (dt, *J* = 5.0, 1.1 Hz, 1H), 7.87 (d, *J* = 2.9 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 14.21, 19.70, 51.41, 61.47, 114.17, 115.07, 125.64, 122.46, 125.97, 125.77, 126.60, 134.22, 136.63, 142.81, 171.17; HR-MS (ESI^+) *m/z* Calcd for $C_{16}H_{17}N_3O_2S$ $[M+H]^+$ 316.11142; Found 316.11105.

Ethyl 2-((2-(5-bromopyridin-2-yl)imidazo[1,2-*a*]pyridin-3-yl)amino)acetate (4s): Brown solid, mp 91.7-93.5 °C; ^1H NMR (500 MHz, $CDCl_3$) δ 1.21 (t, *J* = 7.2 Hz, 3H), 3.88 (s, 2H), 4.17 (q, *J* = 7.2 Hz, 2H), 6.26 (br, 1H, NH), 6.84 (t, *J* = 6.7 Hz, 1H), 7.22-7.14 (m, 1H), 7.60 (d, *J* = 9.1 Hz, 1H), 7.88 (dd, *J* = 8.5, 2.4 Hz, 1H), 8.11 (t, *J* = 8.5 Hz, 2H), 8.64 (d, *J* = 2.3 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 14.17, 48.61, 61.38, 112.78, 117.67, 118.35, 121.97, 122.94, 124.96, 130.57, 139.46, 140.72, 149.91, 170.73; HR-MS (ESI^+) *m/z* Calcd for $C_{16}H_{15}BrN_4O_2$ $[M+H]^+$ 375.04511; Found 375.04483.

Ethyl 2-((8-methyl-2-(5-methylthiophen-2-yl)imidazo[1,2-*a*]pyridin-3-yl)amino)acetate (4t): Brown sticky liquid; ^1H NMR (500 MHz, $CDCl_3$) δ 1.29 (t, *J* = 7.1 Hz, 3H), 2.50 (s, 3H), 2.87 (s, 3H), 3.57 (t, *J* = 5.1 Hz, 1H), 3.83 (d, *J* = 5.2 Hz, 2H), 4.26 (q, *J* = 7.1 Hz, 2H), 6.41 (d, *J* = 6.8 Hz, 1H), 6.74 (d, *J* = 3.0 Hz, 1H), 6.99 (dd, *J* = 8.8, 7.0 Hz, 1H), 7.36 (t, *J* = 6.1 Hz, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 4.26, 15.44, 15.45, 19.43, 51.31, 61.41, 113.55, 115.54, 124.20, 124.36, 124.87, 126.04, 134.38, 135.01, 136.09, 139.88, 143.54, 171.05; HR-MS (ESI^+) *m/z* Calcd for $C_{17}H_{19}N_3O_2S$ $[M+H]^+$ 330.12707; Found 330.12665.

Ethyl 2-((8-methyl-2-(5-methylfuran-2-yl)imidazo[1,2-*a*]pyridin-3-yl)amino)acetate (4u): Brown sticky liquid; ^1H NMR (500 MHz, $CDCl_3$) δ ^1H NMR (500 MHz, $CDCl_3$) δ 1.26 (t, *J* = 7.2 Hz, 3H), 2.38 (s, 3H), 2.59 (s, 3H), 3.84 (d, *J* = 5.7 Hz, 2H), 4.00 (t, *J* = 5.7 Hz, 1H), 4.21 (q, *J* = 7.2 Hz, 2H), 6.09 (dd, *J* = 2.1, 0.9 Hz, 1H), 6.70 (t, *J* = 6.8 Hz, 1H), 6.74 (d, *J* = Hz, 2H), 6.92 (dd, *J* = 6.8, 1.0 Hz, 1H), 8.04 (dd,

J = 6.8, 0.5 Hz, 1H). ^{13}C NMR (125 MHz, $CDCl_3$) δ 13.89, 14.24, 16.78, 49.82, 51.31, 61.36, 107.57, 107.90, 108.70, 111.96, 120.57, 123.17, 127.15, 142.36, 147.94, 151.96, 171.56; HR-MS (ESI$^+$) *m/z* Calcd for $C_{17}H_{19}N_3O_3$ $[M+H]^+$ 319.14992; Found 319.14951.

Ethyl 2-((2-(2-chlorophenyl)-5-methylimidazo[1,2-a]pyridin-3-yl)amino)acetate (4v): Light brown sticky liquid; ^{1}H NMR (500 MHz, $CDCl_3$) δ 1.15 (t, *J* = 7.1 Hz, 3H), 2.94 (s, 3H), 3.49 (d, *J* = 5.7 Hz, 2H), 3.69 (t, *J* = 5.7 Hz, 1H), 4.01 (q, *J* = 7.1 Hz, 2H), 6.48 (d, *J* = 6.7 Hz, 1H) 7.04 (dd, *J* = 8.8, 6.9 Hz, 1H), 7.32 (dd, *J* = 6.2, 2.8 Hz, 2H), 7.39 (d, *J* = 9.0 Hz, 1H), 7.58 (dd, *J* = 5.9, 3.3 Hz, 1H), 7.48-7.45 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 14.14, 19.94, 52.32, 61.10, 113.34, 113.64, 115.97, 124.66, 126.95, 129.58, 129.61, 132.43, 133.34, 133.72, 136.75, 137.10, 143.35, 170.89; HR-MS (ESI$^+$) m/z Calcd for $C_{18}H_{18}ClN_3O_2$ $[M+H]^+$ 344.11603; Found 344.11572.

Ethyl 2-((2-(2-fluorophenyl)-5-methylimidazo[1,2-*a*]pyridin-3-yl)amino)acetate (4w): Light brown sticky liquid; ^{1}H NMR (500 MHz, $CDCl_3$) δ 1.17 (t, *J* = 7.1 Hz, 3H), 2.95 (s, 3H), 3.57 (d, *J* = 5.8 Hz, 2H), 3.81 (br, 1H), 4.04 (q, *J* = 7.2 Hz, 2H), 6.48 (d, *J* = 6.8 Hz, 1H), 7.04 (dd, *J* = 8.9, 6.9 Hz, 1H), 7.17 (dd, *J* = 10.5, 8.6 Hz, 1H), 7.24-7.27 (m, 1H), 7.33-7.38 (m, 1H), 7.42 (d, *J* = 9.0 Hz, 1H), 7.79 (td, *J* = 7.6, 1.7 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 14.16, 19.87, 52.45, 61.12, 113.66, 115.72, 115.91, 124.62, 124.72, 129.74, 129.81, 131.72, 131.75, 133.88, 136.78, 143.83, 170.93; HR-MS (ESI$^+$) *m/z* Calcd for $C_{18}H_{18}FN_3O_2$ $[M+H]^+$ 328.14558; Found 328.14526.

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