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Preparation of 1,2,5-Trisubstituted 1*H*-Imidazoles from Ketenimines and Propargylic Amines by Silver-Catalyzed or Iodine-Promoted Electrophilic Cyclization Reaction of Alkynes

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From readily available propargylic amines, 1,2,5-trisubstituted imidazoles are efficiently obtained through a cascade reaction catalyzed by AgOTf or promoted by molecular iodine. The AgOTf-catalyzed reaction involves nucleophilic addition of propargylic amine to ketenimine, a silver-catalyzed electrophilic cyclization reaction of alkyne, and a tautomerism/isomerism/metal-H exchange cascade. The iodine-mediated counterpart yields 5-formyl-1,2-disubtituted imidazoles, which presumably includes a cascade hydrolysis/oxidation reaction. Furthermore, the presented protocol can be scaled up and the resultant 1,2,5-trisubstituted imidazole can be converted into fused indeno[1,2-*d*]imidazole.

Introduction

Imidazole and its derivatives are an important class of heterocyclic compounds as a result of their prevalence in bioactive natural products[1] and drugs (Figure 1).[2] A tremendous number of imidazoles are valuable ligands in metalloenzymes[3]or precursors of stable carbene ligands.[4] By modification into ionic derivatives, imidazolium salts can be used as environmentally benign solvents[5] and receptors for the recognition of anions.[6] Despite the endeavors to prepare and functionalize this heterocyclic system,[7] it is still challenging to readily achieve high efficiency and regioselectivity.[8]

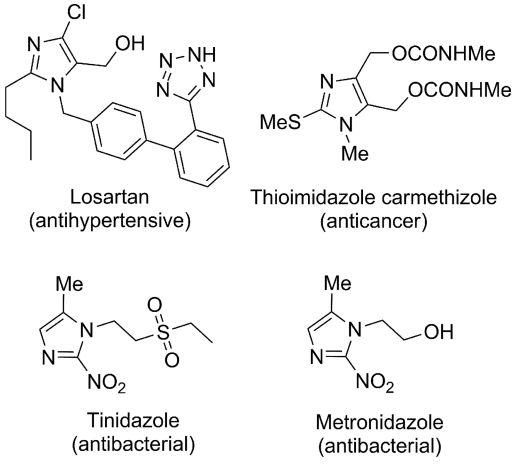

Figure 1. Representative drugs with an imidazole moiety.

Ketenimines are essential intermediates in organic synthesis. Recent progress in the preparation and synthetic utility of ketenimines has been carefully reviewed.[9] In general, there are two categories of ketenimines based on their reactivity and availability. Stable ketenimines[9d] can be fully isolated and characterized, whereas the relatively unstable ones can only be generated in situ as intermediates.[9a–9c] Moreover, propargylic amines have been reported as versatile starting materials in the preparation of various heterocyclic compounds,[10] typically in the Lewis acid catalyzed electrophilic cyclization reaction of alkynes.[11] As a part of our ongoing research on ketenimine chemistry,[9a,9c,10] and encouraged by successes in activating triple bond with silver salts,[12] we became interested in the silver-catalyzed reaction of stable ketenimines and propargylic amines for the one-pot synthesis of 1,2,5-trisubstituted 1*H*-imidazoles. Herein we report the results of this effort.

Results and Discussion

Our initial investigation centered on the reaction between *N*-(2,2-diphenylvinylidene)aniline (**1a**) and prop-2-yn-1-amine (**2a**) as the model reaction (Table 1). When the reaction was performed in the presence of CuI (0.1 equiv.) and triethylamine (Et_3N; 1.0 equiv.) in tetrahydrofuran (THF) at reflux temperatures for 3 h, 2-benzhydryl-5-methyl-1-phenyl-1*H*-imidazole (**3a**) was obtained in 67% isolated yield (Table 1, Entry 1). The structure of **3a** was confirmed by single crystal analysis of its analogues **3d**.[13] Further optimization studies revealed that the absence of the catalyst decreased the yield of **3a** to 38% (Table 1, Entry 2). By screening the catalyst, we discovered that AgOTf was opti-

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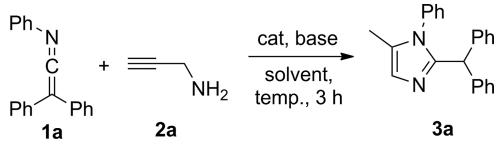
mal among CuI, CuBr, CuCl, CuOTf, and $Cu(OTf)_2$ (Table 1, Entries 1 and 3–7). THF was screened as the optimal solvent among all examined solvents, including dichloroethane (DCE), acetonitrile, toluene, and *N*,*N*-dimethylformamide (DMF) (Table 1, Entries 7–11). Furthermore, a suitable base additive was essential to this transformation. The use of other bases, such as K_2CO_3, Cs_2CO_3, and 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU), decreased the yield (Table 1, Entries 12–14). By decreasing the amount of Et_3N (from 1.0 to 0.5 equiv.) or by eliminating the base, a poor yield was observed (Table 1, Entries 15 and 16). Attempts to carry out the reaction at a lower temperature and for a short reaction time both led to a decrease in the yield (Table 1, Entries 17–19). Accordingly, the optimal reaction conditions were established to obtain **3a** in 80% yield by heating to reflux temperatures a mixture of **1a** (1 mmol), **2a** (1 mmol), AgOTf (0.1 mmol), and Et_3N (1 mmol) in THF (10 mL) for 3 h (Table 1, Entry 7).

Table 1. Screening of the reaction conditions for the formation of **3a**.



1a **2a** **3a**

Entry[a]	Catalyst	Solvent	Base/equiv.	Temp [°C]	Yield [%][b]
1	CuI	THF	Et_3N/1	reflux	67
2	none	THF	Et_3N/1	reflux	38
3	CuBr	THF	Et_3N/1	reflux	59
4	CuCl	THF	Et_3N/1	reflux	60
5	CuOTf	THF	Et_3N/1	reflux	62
6	$Cu(OTf)_2$	THF	Et_3N/1	reflux	57
7	AgOTf	THF	Et_3N/1	reflux	80
8	AgOTf	DCE	Et_3N/1	80	54
9	AgOTf	MeCN	Et_3N/1	80	71
10	AgOTf	toluene	Et_3N/1	80	55
11	AgOTf	DMF	Et_3N/1	80	69
12	AgOTf	THF	K_2CO_3/1	reflux	64
13	AgOTf	THF	Cs_2CO_3/1	reflux	50
14	AgOTf	THF	DBU/1	reflux	35
15	AgOTf	THF	Et_3N/0.5	reflux	46
16	AgOTf	THF	none	reflux	40
17	AgOTf	THF	Et_3N/1	50	54
18	AgOTf	THF	Et_3N/1	reflux	78[c]
19	AgOTf	THF	Et_3N/1	reflux	63[d]

[a] Reaction conditions: **1a** (1 mmol), **2a** (1 mmol), cat (0.1 equiv.), solvent (10 mL), 3 h. [b] Isolated yield referred to **1a**. [c] The reaction time is 2 h. [d] The reaction time is 1 h.

Next, we investigated the substrate scope and the results are summarized in Table 2. Firstly, substituent R^2 on the nitrogen of ketenimines **1** was screened for the reaction of **1** with **2a**. When the R^2 group was altered from phenyl (**1a**) to *p*-tolyl (**1b**), *p*-methoxyphenyl (**1c**), *p*-bromophenyl (**1d**), *p*-nitrophenyl (**1e**), *p*-ethoxycarbonylphenyl (**1f**), and *m*-nitrophenyl (**1i**), corresponding products **3a**–**3f** and **3i** were isolated in good yields (62–84%) and no apparent electronic effect was observed. Whereas, when two ethyl groups (**1g**) or a bromo (**1h**) occupied the *ortho* position of phenyl of R^2, desired products **3g** and **3h** were isolated in slightly lower yields owing to the steric hindrance of the R^2 group. Besides the substituted phenyl, the R^2 group could be heterocyclic, such as 3-pyridinyl (**1j**). In this case, **3j** was obtained in 69% yield. Then, the R^1 group on ketenimines **1** was changed from phenyl to methyl (**1k**–**1o**). In these cases, corresponding products **3k**–**3o** were prepared in yields ranging from 66% to 71%. Finally, we tested the R^3 group on propargylic amines **2**. A series of aryl-substituted propargylic amines **2b**–**2g** could react with *N*-(2,2-diphenylvinylidene)-4-methylaniline (**1b**) to afford corresponding products **3p**–**3u** in yields ranging 53–71%, which were lower than that obtained from the reaction between **1b** and **2a**.

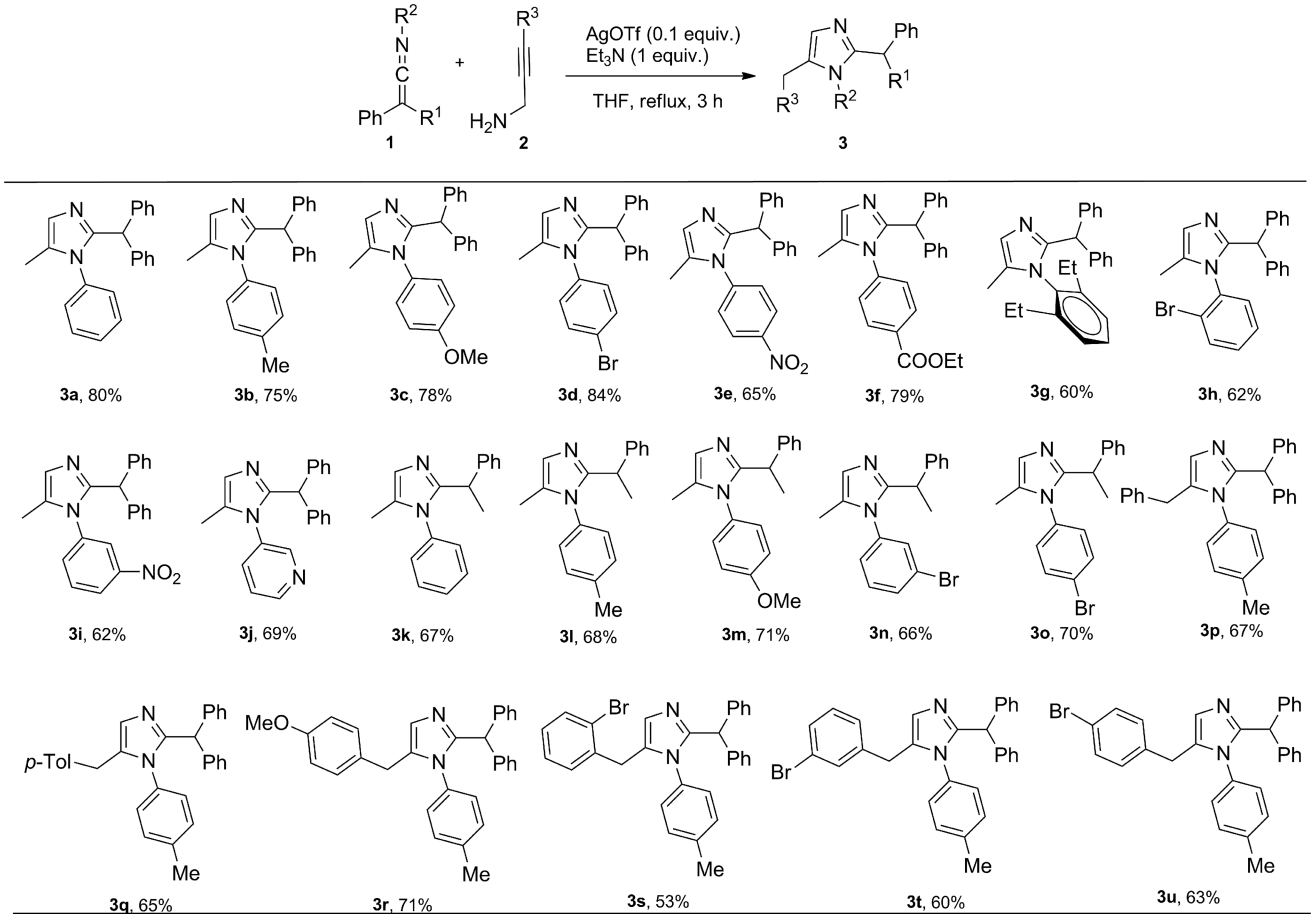
Our method can be operated on gram scale. For instance, **3b** (1.764 g) was isolated when the mixture of *N*-(2,2-diphenylvinylidene)-4-methylaniline (**1b**, 1.981 g), **2a** (0.385 g), AgOTf (0.179 g), and Et_3N (0.707 g) in 30 mL of THF was heated to reflux temperatures for 3 h.

Ethoxycarbonyl-substituted ketenimines **4** were also evaluated for this transformation under the established reaction conditions and the results are summarized in Table 3. When the R group on **4** was an aryl group, such as phenyl (**4a**), *p*-chlorophenyl (**4b**), *m*-chlorophenyl (**4c**), and *o*-chlorophenyl (**4d**), imidazoles **5a**–**5d** were obtained in moderate yields. Notably, an oxidized product **6d** was isolated in 10% yield along with **5d** in the last case. Furthermore, the R group on **4** could be alkyl groups, such as cyclohexyl (**4e**), isopropyl (**4f**), ethyl (**4g**), and *tert*-butyl (**4h**). In these cases, corresponding products **5e**–**5h** were isolated in 54–96% yield.

A possible mechanism for the formation of imidazoles **3** and **5** is postulated in Scheme 1. First, the nucleophilic addition of propargylic amine **2a** to ketenimine **1a** forms intermediate **A**. In the presence of a Lewis acid, the triple bond is activated and becomes electron deficient. Intramolecular attack of nitrogen on the electron-deficient triple bond forms intermediate **B** through a silver(I)-catalyzed 5-*exo*-*dig* cyclization.[12] Subsequent metal-H exchange and a 1,3-H shift generate **3a**.

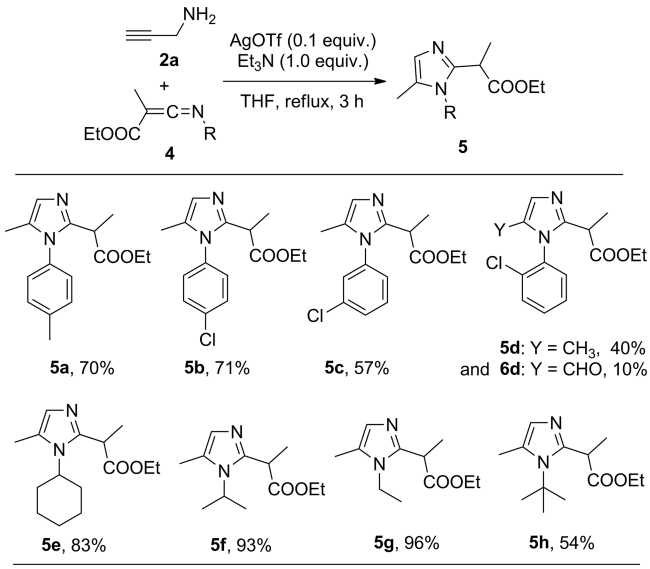
Attracted by the formation of aforementioned **6d** as shown in Table 3, we optimized the reaction conditions for this transformation based on the model reaction between **4a** and **2a** (Table 4). Assisted by the participation of iodine, aldehyde **6a** was obtained in 30% yield under an air atmosphere (Table 4, Entry 1). The yield of **6a** was improved to 48% under oxygen (Table 4, Entry 2). By switching the iodine source to *N*-iodosuccinimide (NIS) decreased the yield to 41% (Table 4, Entry 3). By reducing the amount of iodine to 0.5 equiv., the yield of **6a** approached 55% (Table 4, Entry 4). Interestingly, **6a** could be obtained in moderate yield in the absence of AgOTf (Table 4, Entries 5–7). Attempts to carry out the reaction at room temperature led to a significant increase in yield (Table 4, Entry 8). Molecule oxygen was essential to have a better transformation relative to reactions conducted in air or N_2 (Table 4, Entries 9 and 10). However, by adding a supplemental oxidant, such as hydrogen peroxide or *tert*-butyl hydrogen peroxide, did not improve the yield (Table 4, Entries 11 and 12).

Table 2. Preparation of **3a**–**3u**.[a]

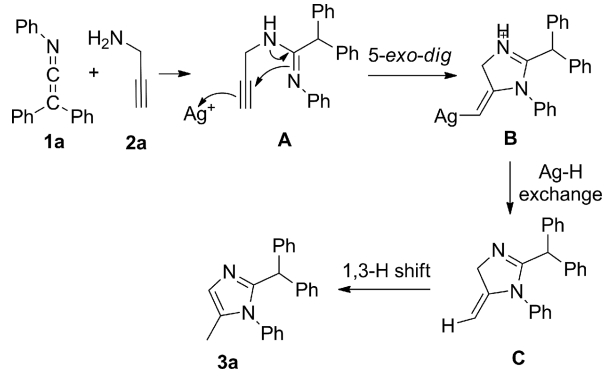


[a] Reaction conditions: **1** (1 mmol), **2** (1 mmol), AgOTf (0.1 mmol), Et_3N (1 mmol), THF (10 mL), reflux, 3 h; Isolated yield refers to **1**.

Table 3. Preparation of **5a**–**5h**.[a]



[a] Reaction conditions: **1** (1 mmol), **2** (1 mmol), AgOTf (0.1 mmol), Et_3N (1 mmol), THF (10 mL), reflux, 3 h; Isolated yield refers to **1**.

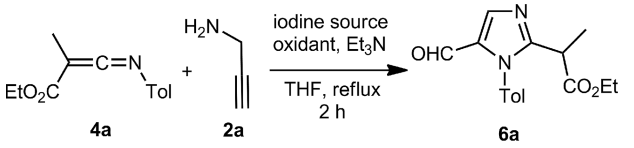


Scheme 1. Proposed mechanism for the formation of **3a**.

Therefore, the optimal reaction condition for preparation of **6a** was established. A mixture of **4a** (0.5 mmol), **2a** (0.5 mmol), I_2 (2 mmol), and Et_3N (0.5 mmol) in 2 mL of THF was allowed to react under O_2 at room temperature for 2 h (Table 4, Entry 8).

With the optimized reaction conditions in hand, we synthesized aldehydes **6a**–**6g** in 40–74% yields (Table 5, Condi-

Table 4. Screening of reaction conditions for the formation of **6a**.[a]



Entry	Catalyst	I^+ source [equiv.]	Atmosphere	Yield [%][b]
1	AgOTf	I_2 (1)	air	30
2	AgOTf	I_2 (1)	O_2	48
3	AgOTf	NIS (1)	O_2	41
4	AgOTf	I_2 (0.5)	O_2	55
5	–	I_2 (0.5)	O_2	42
6	–	I_2 (1)	O_2	45
7	–	I_2 (2)	O_2	47
8	–	I_2 (2)	O_2	71[c]
9	–	I_2 (2)	air	60[c]
10	–	I_2 (2)	N_2	27[c]
11	–	I_2 (2)	air	66[c,d]
12	–	I_2 (2)	air	60[c,e]

[a] Reaction condition: **4a** (0.5 mmol), **2a** (0.5 mmol), Et_3N (0.5 mmol), THF (2 mL), reflux, 2 h. [b] Isolated yield. [c] Room temperature. [d] 30% H_2O_2 (2 equiv.) was used. [e] *tert*-Butyl hydroperoxide (2 equiv.) was used.

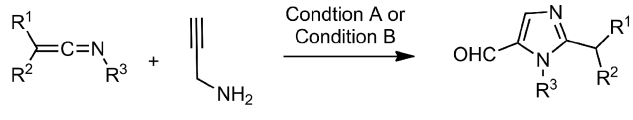
tion A). For cases in which R^1 and/or R^2 were aromatic groups, we obtained a complex mixture. To have a clear transformation, we mixed ketenimine **4** with propargylic amine **2a** at room temperature for 30 min. After propargylic amidine formed, as confirmed by TLC, iodine was added to allow further reaction at room temperature for 1 h. In this way, aldehydes **6h**–**6l** were prepared in yields ranging 46–75% (Table 5, Condition B).

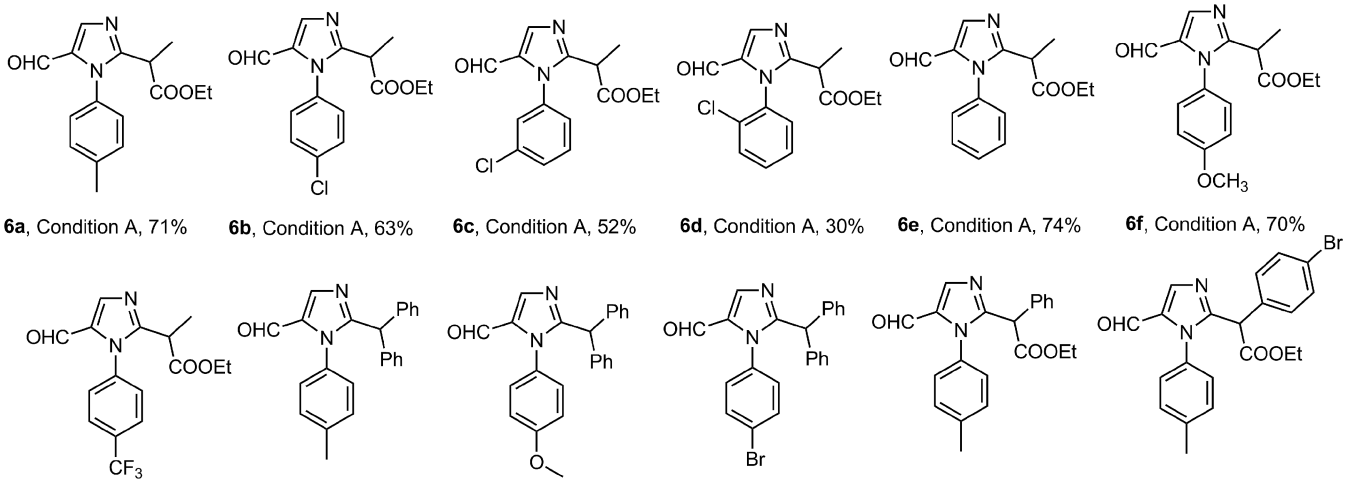
To gain a better insight into the mechanism, we subjected **5a** and **3a** to reactions under Conditions A and B in Table 5, respectively. However, corresponding oxidized products **6a** and **6h** were not obtained with the recovery of the starting materials. In this case, the methyl groups on **5a** and **3a** could not be oxidized even if more hydrogen peroxide was used.[11a] This result indicated that **3a** and **5a** were not the intermediates in the formation of **6h** and **6a**, respectively.

Based on these results, we postulated a working mechanism for the formation of **6a** (Scheme 2). The initial step is similar to that of the formation of **3**. Propargylic amidine **A** is generated by nucleophilic addition of propargylic amine **2a** to ketenimine **4a**. Subsequently, **A** undergoes iodine-mediated electrophilic cyclization reaction to form vinyl iodide **B**,[11b] which can aromatize to form **C**. Finally, **C** is immediately oxidized to aldehyde **6a** via a cascade hydrolysis/oxidation[11b] or through a free radical process.[11c,11e]

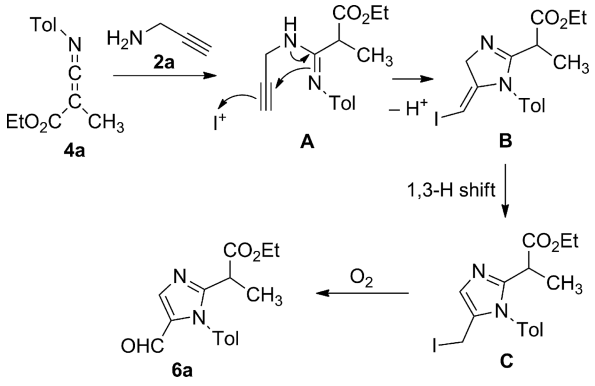
Importantly, the resulted functionalized products could be extended for the synthesis of more complicated heterocyclic systems. For example, **3s** was converted into 2-benzhydryl-1-*p*-tolyl-1,8-dihydroindeno[2,1-*d*]imidazole (**7**) in 55% yield through an intramolecular Heck reaction (Scheme 3).

Table 5. Preparation of aldehydes **6a**–**6l**.[a]

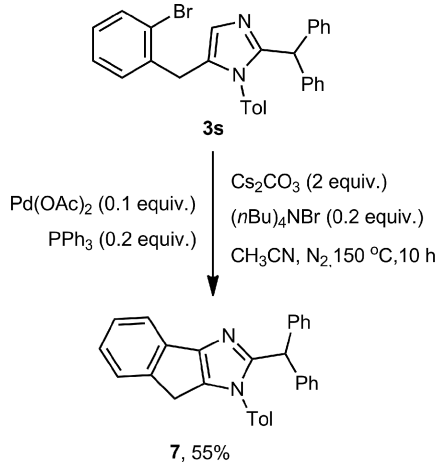




[a] Condition A: **4** (0.5 mmol), **2a** (0.5 mmol), Et_3N (0.5 mmol), I_2 (1 mmol), O_2 (balloon), THF (2 mL), room temp., 2 h; Condition B: (1) **4** (0.5 mmol), **2a** (0.5 mmol), Et_3N (0.5 mmol), THF (2 mL), room temp., 30 min; (2) I_2 (1 mmol), 30% H_2O_2 (1 mmol), room temp., 1 h; Isolated yield refers to **4**.



Scheme 2. Proposed mechanism for the formation of **6**.



Scheme 3. Synthesis of dihydroindenoimidazole **7**.

Conclusions

We developed an efficient and feasible method for the synthesis of 1,2,5-trisubstituted imidazoles from readily available propargylic amines and ketenimines. The cascade reaction was catalyzed by AgOTf and a typical silver-catalyzed electrophilic cyclization reaction of alkynes was presented. This protocol can be scaled up and the resultant 1,2,5-trisubstituted imidazole converted into fused indeno[1,2-*d*]imidazole by rational design of the structure of the propargylic amine substrate. Moreover, by using iodine as a Lewis acid, the cascade reaction between propargylic amines and ketenimines furnished 5-formyl-1,2-disubstituted imidazoles in moderate to good yields.

Experimental Section

General: Unless otherwise mentioned, solvents and reagents were purchased from commercial sources and used as received. THF and toluene were distilled from Na before use. MeCN and DCE was distilled from CaH_2. Melting points were measured with a micro melting point apparatus. Infrared spectra were obtained with an FTIR spectrometer. ^{1}H NMR spectra were recorded with a 500 or 400 MHz spectrometer in $CDCl_3$ solution and the chemical shifts were reported relative to internal standard tetramethylsilane (δ = 0 ppm). The following abbreviations are used to describe peak patterns as appropriate: s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet. ^{13}C NMR were recorded at 125 or 100 MHz and referenced to the internal solvent signals (central peak is 77.00 ppm). High-resolution mass spectra (HRMS) data were obtained with an electron ionization time-of-flight (EI-TOF) mass spectrometer.

General Procedure for the Synthesis of Imidazoles 3 or 5: To a mixture of AgOTf (0.1 mmol), ketenimine **1** or **4** (1 mmol), and Et_3N (1 mmol) in THF (10 mL) was added propargylic amine **2** (1 mmol). The mixture was stirred at reflux temperatures for 3 h and then evaporated in vacuo. The residue was subjected to silica gel column chromatography with petroleum ether/ethyl acetate as eluent.

2-Benzhydryl-5-methyl-1-phenyl-1*H*-imidazole (3a): White solid (260 mg, 0.80 mmol, 80%), m.p. 141–142 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.43–7.40 (m, 3 H), 7.26–7.22 (m, 4 H), 7.19–7.14 (m, 6 H), 7.01–6.99 (m, 2 H), 6.90 (s, 1 H), 5.12 (s, 1 H), 2.01 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 148.7, 141.6, 136.3, 129.3, 128.8, 128.8, 128.3, 128.2, 128.0, 126.5, 125.6, 49.3, 10.0 ppm. IR (film): $\tilde{\nu}$ = 3060, 3026, 2920, 1598, 1498, 1452, 1413, 774, 696 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{23}H_{20}N_2$ (M^+) 324.1626; found 324.1617.

2-Benzhydryl-5-methyl-1-(p-tolyl)-1*H*-imidazole (3b): White solid (255 mg, 0.75 mmol, 75%), m.p. 103–104 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.25–7.15 (m, 12 H), 6.89–6.87 (m, 3 H),5.12 (s, 1 H), 2.40 (s, 3 H), 1.96 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 148.7, 141.8, 138.8, 133.5, 129.9, 128.8, 128.2, 128.2, 127.6, 126.4, 125.5, 49.1, 21.1, 10.0 ppm. IR (film): $\tilde{\nu}$ = 3060, 3026, 2921, 1600, 1516, 1494, 1453, 1416, 1203, 1145, 1014, 823, 724, 701 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{24}H_{22}N_2$ (M^+) 338.1783; found 338.1783.

2-Benzhydryl-1-(4-methoxyphenyl)-5-methyl-1*H*-imidazole (3c): White solid (273 mg, 0.78 mmol, 78%), m.p. 96–97 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.26–7.22 (m, 4 H), 7.19–7.15 (m, 6 H), 6.90–6.88 (m, 5 H), 5.12 (s, 1 H), 3.85 (s, 3 H), 1.95 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 159.7, 149.0, 141.7, 129.0, 128.8, 128.4, 128.3, 126.5, 125.4, 114.5, 55.5, 49.2, 10.0 ppm. IR (film): $\tilde{\nu}$ = 3059, 3025, 2919, 1600, 1515, 1494, 1452, 1250, 1144, 823, 753, 700 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{24}H_{22}N_2O$ (M^+) 354.1732; found 354.1740.

2-Benzhydryl-1-(4-bromophenyl)-5-methyl-1*H*-imidazole (3d): White solid (336 mg, 0.84 mmol, 84%), m.p. 165–166 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.52 (d, *J* = 8.0 Hz, 2 H), 7.26–7.23 (m, 4 H), 7.20–7.14 (m, 6 H), 6.90 (s, 1 H), 6.86 (d, *J* = 8.0 Hz, 2 H), 5.09 (s, 1 H), 1.96 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 148.6, 141.3, 135.3, 132.6, 129.6, 128.7, 128.4, 128.1, 126.6, 125.9, 122.9, 49.4, 10.0 ppm. IR (film): $\tilde{\nu}$ = 3085, 3060, 3025, 2919, 1492, 1451, 1412, 1069, 1009, 701 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{23}H_{19}BrN_2$ (M^+) 402.0732; found 402.0736.

2-Benzhydryl-5-methyl-1-(4-nitrophenyl)-1*H*-imidazole (3e): White solid (241 mg, 0.65 mmol, 65%), m.p. 180–181 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.26 (d, *J* = 8.8 Hz, 2 H), 7.27–7.12 (m, 12 H), 6.94 (s, 1 H), 5.09 (s, 1 H), 1.99 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 148.6, 147.7, 142.0, 140.9, 129.0, 128.7, 128.5, 128.1, 126.9, 126.6, 124.7, 49.8, 10.1 ppm. IR (film): $\tilde{\nu}$ = 3084, 3026, 2920, 1597, 1525, 1495, 1452, 1407, 1342, 1106, 858, 732, 703 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{23}H_{19}N_3O_2$ (M^+) 369.1477; found 369.1475.

Ethyl 4-(2-Benzhydryl-5-methyl-1*H*-imidazol-1-yl)benzoate (3f): Yellow oil (313 mg, 0.79 mmol, 79%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.09 (d, *J* = 8.4 Hz, 2 H), 7.24–7.12 (m, 10 H), 7.08

(d, J = 8.4 Hz, 2 H), 6.91 (s, 1 H), 5.11 (s, 1 H), 4.02 (q, J = 7.2 Hz, 2 H), 1.97 (s, 3 H), 1.42 (t, J = 7.2 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 165.5, 148.5, 141.2, 140.1, 131.0, 130.7, 128.8, 128.4, 128.1, 128.0, 126.7, 125.9, 61.4, 49.4, 14.3, 10.0 ppm. IR (film): $\tilde{\nu}$ = 3061, 3026, 2980, 1719, 1608, 1494, 1452, 1411, 1275, 1102, 1013, 702 cm^{-1}. HRMS (EI): m/z calcd. for $C_{26}H_{24}N_2O_2$ (M^+) 396.1838; found 396.1844.

2-Benzhydryl-1-(2,6-diethylphenyl)-5-methyl-1*H*-imidazole(3g): Yellow oil (228 mg, 0.60 mmol, 60%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.43–7.38 (m, 3 H),7.22–7.17 (m, 10 H), 6.98 (s, 1 H), 4.80 (s, 1 H), 2.00–1.93 (m, 2 H), 1.87 (s, 3 H), 1.86–1.78 (m, 2 H), 0.92 (t, J = 7.6 Hz, 6 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 148.3, 142.0, 140.9, 132.6, 129.6, 128.8, 128.4, 127.8, 126.6, 126.1, 125.9, 49.3, 23.0, 13.5, 9.7 ppm. IR (film): $\tilde{\nu}$ = 3059, 3026, 2969, 2934, 2876, 1598, 1494, 1466, 1436, 1411, 1032, 770, 732, 697 cm^{-1}. HRMS (EI): m/z calcd. for $C_{27}H_{28}N_2$ (M^+) 380.2252; found 380.2248.

2-Benzhydryl-1-(2-bromophenyl)-5-methyl-1*H*-imidazole (3h): Yellow oil (245 mg, 0.62 mmol, 62%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.75–7.73 (m, 1 H),7.33–7.16 (m, 10 H),7.06–7.05 (m, 2 H), 6.92 (s, 1 H), 6.78–6.76 (m, 1 H), 4.96 (s, 1 H), 1.93 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 148.5, 141.2, 141.0, 135.6, 133.5, 130.7, 129.1, 128.8, 128.4, 128.2, 126.6, 125.8, 123.4, 49.8, 9.8 ppm. IR (film): $\tilde{\nu}$ = 3060, 3026, 2918, 1641, 1483, 1452, 1409, 1031, 768, 731, 700 cm^{-1}. HRMS (EI): m/z calcd. for $C_{23}H_{19}BrN_2$ (M^+) 402.0732; found 402.0736.

2-Benzhydryl-5-methyl-1-(3-nitrophenyl)-1*H*-imidazole (3i): White solid (229 mg, 0.62 mmol, 62%), m.p. 129–130 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.28 (d, J = 10.4 Hz, 1 H), 7.82–7.81 (m, 1 H),7.57 (t, J = 8.0 Hz, 1 H), 7.32–7.17 (m, 7 H), 7.12–7.10 (m, 4 H), 6.93 (s, 1 H), 5.08 (s,1 H), 1.99 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 148.8, 148.5, 140.7, 137.5, 134.0, 130.2, 128.7, 128.5, 128.3, 126.9, 126.2, 123.7, 123.3, 49.9, 10.0 ppm. IR (film): $\tilde{\nu}$ = 3086, 3026, 1534, 1493, 1452, 1406, 1350, 747, 702 cm^{-1}. HRMS (EI): m/z calcd. for $C_{23}H_{19}N_3O_2$ (M^+) 369.1477; found 369.1474.

3-(2-Benzhydryl-5-methyl-1*H*-imidazol-1-yl)pyridine (3j): White solid (223 mg, 0.69 mmol, 69%), m.p. 169–170 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.67 (s, 1 H), 8.33 (s, 1 H),7.32–7.31 (m, 1 H),7.25–7.19 (m, 7 H),7.13–7.12 (m, 4 H), 6.92 (s, 1 H), 5.08 (s, 1 H), 1.98 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 150.0, 149.0, 148.9, 141.0, 135.4, 133.1, 128.8, 128.5, 126.8, 126.2, 123.8, 49.7, 10.0 ppm. IR (film): $\tilde{\nu}$ = 3059, 3026, 2920, 2934, 1600, 1486, 1452, 1427, 1206, 1078, 1031, 1009, 813, 733, 701 cm^{-1}. HRMS (EI): m/z calcd. for $C_{22}H_{19}N_3$ (M^+) 325.1579; found 325.1581.

5-Methyl-1-phenyl-2-(1-phenylethyl)-1*H*-imidazole (3k): Yellow oil (176 mg, 0.67 mmol, 67%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.50–7.00 (m, 7 H), 6.92 (dd, J = 7.8, 1.4 Hz, 2 H), 6.86 (d, J = 0.9 Hz, 1 H), 6.74–6.36 (br., 1 H), 3.85 (q, J = 7.2 Hz, 1 H), 1.92 (s, 3 H), 1.65 (d, J = 7.2 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 150.4, 144.3, 136.3, 129.1, 128.55, 128.51, 128.2, 127.2, 126.1, 124.7, 38.3, 21.6, 9.9 ppm. IR (film): $\tilde{\nu}$ = 3059, 3026, 2918, 1641, 1494, 1452, 1427, 1031, 731, 702 cm^{-1}. HRMS (EI): m/z calcd. for $C_{18}H_{18}N_2$ (M^+) 262.1470; found 262.1469.

5-Methyl-2-(1-phenylethyl)-1-(*p*-tolyl)-1*H*-imidazole (3l): Yellow oil (188 mg, 0.68 mmol, 68%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.35–7.00 (m, 6 H), 7.00–6.91 (m, 2 H), 6.85 (d, J = 0.9 Hz, 1 H), 6.64–6.28 (br., 1 H), 3.84 (q, J = 7.2 Hz, 1 H), 2.39 (s, 3 H), 1.92 (d, J = 0.9 Hz, 3 H), 1.64 (d, J = 7.2 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 150.5, 144.4, 138.6, 133.6, 129.7, 128.5, 128.2, 127.2, 126.1, 124.6, 38.2, 21.7, 21.1, 9.9 ppm. IR (film): $\tilde{\nu}$ = 3027, 2976, 2925, 2360, 2340, 1517, 1492, 1453, 1415, 825, 700 cm^{-1}. HRMS (EI): m/z calcd. for $C_{19}H_{20}N_2$ (M^+) 276.1626; found 276.1626.

1-(4-Methoxyphenyl)-5-methyl-2-(1-phenylethyl)-1*H*-imidazole(3m): Yellow oil (207 mg, 0.71 mmol, 71%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.25–7.03 (m, 4 H), 7.02–6.87 (m, 3 H), 6.84 (d, J = 0.7 Hz, 1 H), 6.81–6.60 (br., 1 H), 6.55–6.32 (br., 1 H), 3.87–3.82 (m, 4 H), 1.91 (d, J = 0.6 Hz, 3 H), 1.64 (d, J = 7.2 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 159.5, 150.7, 144.4, 128.81, 128.75, 128.2, 127.2, 126.1, 124.5, 114.2, 55.4, 38.3, 21.6, 9.9 ppm. IR (film): $\tilde{\nu}$ = 2974, 2928, 1608, 1515, 1493, 1453, 1250, 838, 700 cm^{-1}. HRMS (EI): m/z calcd. for $C_{19}H_{20}N_2O$ (M^+) 292.1576; found 292.1577.

1-(3-Bromophenyl)-5-methyl-2-(1-phenylethyl)-1*H*-imidazole (3n): Yellow oil (224 mg, 0.66 mmol, 66%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.54–7.50 (m, 1 H), 7.40–6.99 (m, 5 H), 6.94–6.87 (m, 2 H), 6.85 (d, J = 0.9 Hz, 1 H), 6.58 (s, 1 H), 3.82 (q, J = 6.7 Hz, 1 H), 1.93 (d, J = 0.9 Hz, 3 H), 1.66 (d, J = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 150.4, 144.1, 137.5, 131.8, 130.3, 128.5, 128.4, 127.2, 126.4, 125.0, 122.4, 38.6, 21.5, 9.9 ppm. IR (film): $\tilde{\nu}$ = 2977, 2926, 2362, 2342, 1591, 1482, 1451, 1427, 1403 cm^{-1}. HRMS (EI): m/z calcd. for $C_{18}H_{17}BrN_2$ (M^+) 340.0575; found 340.0573.

1-(4-Bromophenyl)-5-methyl-2-(1-phenylethyl)-1*H*-imidazole (3o): Colorless oil (237 mg, 0.70 mmol, 70%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.70–7.29 (br., 2 H), 7.25–6.97 (m, 4 H), 6.96–6.88 (m, 2 H), 6.86 (d, J = 0.9 Hz, 1 H), 6.70–6.20 (br., 1 H), 3.82 (q, J = 7.2 Hz, 1 H), 1.92 (d, J = 0.9 Hz, 3 H), 1.65 (d, J = 7.2 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 150.3, 144.1, 135.3, 132.3, 129.6, 128.5, 128.3, 127.1, 126.3, 124.9, 122.7, 38.5, 21.5, 9.9 ppm. IR (film): $\tilde{\nu}$ = 2976, 2925, 1492, 1453, 1140, 1069, 1008, 834, 700 cm^{-1}. HRMS (EI): m/z calcd. for $C_{18}H_{17}BrN_2$ (M^+) 340.0575; found 340.0569.

2-Benzhydryl-5-benzyl-1-(*p*-tolyl)-1*H*-imidazole (3p): White solid (276 mg, 0.67 mmol, 67%), m.p. 159–160 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.25–7.21 (m, 4 H),7.18–7.14 (m, 9 H), 7.08 (d, J = 8 Hz, 2 H), 6.93–6.91 (m, 3 H), 6.70 (d, J = 7.6 Hz, 2 H), 5.09 (s, 1 H), 3.66 (s, 2 H), 2.37 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 149.3, 141.7, 138.9, 138.3, 133.2, 131.5, 129.7, 128.8, 128.5, 128.3, 128.1, 128.0, 126.6, 126.5, 126.1, 49.0, 31.0, 21.2 ppm. IR (film): $\tilde{\nu}$ = 3060, 3026, 2920, 1601, 1516, 1494, 1454, 1423, 1031, 822, 751, 696 cm^{-1}. HRMS (EI): m/z calcd. for $C_{30}H_{26}N_2$ (M^+) 414.2096; found 414.2099.

2-Benzhydryl-5-(4-methylbenzyl)-1-(*p*-tolyl)-1*H*-imidazole (3q): White solid (278 mg, 0.65 mmol, 65%), m.p. 165–166 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.24–7.21 (m, 4 H), 7.19–7.12 (m, 6 H), 7.09 (d, J = 8.0 Hz, 2 H), 6.97 (d, J = 7.9 Hz, 2 H), 6.88 (s, 1 H), 6.82 (d, J = 7.9 Hz, 2 H), 6.72 (d, J = 8.2 Hz, 2 H), 5.09 (s, 1 H), 3.60 (s, 2 H), 2.37 (s, 3 H), 2.27 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 149.2, 141.7, 138.8, 135.6, 135.2, 133.2, 131.8, 129.7, 128.8, 128.4, 128.3, 128.0, 126.4, 126.4, 49.0, 30.5, 21.1, 20.9 ppm. IR (film): $\tilde{\nu}$ = 3025, 2920, 1600, 1515, 1493, 1453, 1424, 1304, 1032, 822, 733, 700 cm^{-1}. HRMS (EI): m/z calcd. for $C_{31}H_{28}N_2$ (M^+) 428.2260; found 428.2252.

2-Benzhydryl-5-(4-methoxybenzyl)-1-(*p*-tolyl)-1*H*-imidazole (3r): Yellow solid (315 mg, 0.71 mmol, 71%), m.p. 147–148 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.26–7.20 (m, 4 H), 7.20–7.12 (m, 6 H), 7.09 (d, J = 8.0 Hz, 2 H), 6.88 (s, 1 H), 6.84 (d, J = 8.6 Hz, 2 H), 6.74–6.66 (m, 4 H), 5.09 (s, 1 H), 3.74 (s, 3 H), 3.59 (s, 2 H), 2.38 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 158.0, 149.2, 141.7, 138.8, 133.3, 132.0, 130.4, 129.7, 129.5, 128.8, 128.2, 128.0,

126.5, 126.3, 113.5, 55.2, 49.0, 30.1, 21.2 ppm. IR (film): $\tilde{\nu}$ = 3060, 3027, 2834, 1611, 1514, 1455, 1246, 1175, 1033, 821, 701 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{31}H_{28}N_2O$ (M^+) 444.2202; found 444.2206.

2-Benzhydryl-5-(2-bromobenzyl)-1-(*p*-tolyl)-1*H*-imidazole (3s): Brown solid (261 mg, 0.53 mmol, 53%), m.p. 90–92 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 7.45–7.38 (m, 1 H), 7.28–7.20 (m, 4 H), 7.20–7.14 (m, 6 H), 7.14–7.06 (m, 3 H), 7.02–6.98 (m, 2 H), 6.84 (s, 1 H), 6.77 (d, *J* = 8.2 Hz, 2 H), 5.12 (s, 1 H), 3.77 (s, 2 H), 2.36 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 149.3, 141.6, 139.0, 137.8, 133.0, 132.5, 130.5, 130.1, 129.8, 128.8, 128.2, 127.9, 127.8, 127.2, 127.1, 126.5, 124.3, 49.1, 31.1, 21.1 ppm. IR (film): $\tilde{\nu}$ = 3059, 2025, 2921, 1600, 1516, 1494, 1423, 1026, 823, 751, 701 cm^{-1}. HRMS (EI): *m*/*z* calcd. For $C_{30}H_{25}N_2Br$ (M^+) 492.1201; found 492.1200.

2-Benzhydryl-5-(3-bromobenzyl)-1-(*p*-tolyl)-1*H*-imidazole(3t): White solid (295 mg, 0.60 mmol, 60%), m.p. 130–131 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 7.27–7.21 (m, 5 H), 7.20–7.11 (m, 6 H), 7.09 (d, *J* = 8.0 Hz, 2 H), 7.01 (t, *J* = 7.8 Hz, 1 H), 6.95 (s, 1 H), 6.91 (s, 1 H), 6.86 (d, *J* = 7.6 Hz, 1 H), 6.65 (d, *J* = 8.1 Hz, 2 H), 5.09 (s, 1 H), 3.64 (s, 2 H), 2.39 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 149.7, 141.6, 140.7, 139.1, 133.0, 131.6, 130.8, 129.8, 129.7, 129.3, 128.8, 128.3, 127.9, 127.2, 126.65, 126.55, 122.1, 49.1, 30.7, 21.2 ppm. IR (KBr): $\tilde{\nu}$ = 3064, 3023, 1595, 1515, 1474, 1453, 1427, 1308, 1071, 824, 756, 555 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{30}H_{25}N_2Br$ (M^+) 492.1201; found 492.1207.

2-Benzhydryl-5-(4-bromobenzyl)-1-(*p*-tolyl)-1*H*-imidazole (3u): White solid (310 mg, 0.63 mmol, 63%), m.p. 186–187 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 7.29–7.20 (m, 6 H), 7.20–7.16 (m, 2 H), 7.16–7.11 (m, 4 H), 7.09 (d, *J* = 8.0 Hz, 2 H), 6.91 (s, 1 H), 6.78 (d, *J* = 8.4 Hz, 2 H), 6.68 (d, *J* = 8.2 Hz, 2 H), 5.08 (s, 1 H), 3.61 (s, 2 H), 2.38 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 149.6, 141.5, 139.1, 137.4, 133.0, 131.2, 130.9, 130.2, 129.8, 128.8, 128.3, 127.9, 126.61, 126.55, 120.0, 49.0, 30.4, 21.2 ppm. IR (film): $\tilde{\nu}$ = 3024, 2917, 1600, 1516, 1487, 1453, 1422, 1307, 1071, 1011, 849, 701 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{30}H_{25}N_2Br$ (M^+) 492.1201; found 492.1199.

Ethyl 2-[5-Methyl-1-(*p*-tolyl)-1*H*-imidazol-2-yl]propanoate (5a): Pale yellow oil (190 mg, 0.7 mmol, 70%). ^1H NMR (400 MHz, $CDCl_3$): δ = 7.29 (d, *J* = 8.3 Hz, 2 H), 7.10 (br., 2 H), 6.83 (s, 1 H), 4.12–3.97 (m, 2 H), 3.59 (q, *J* = 7.2 Hz, 1 H), 2.43 (s, 3 H), 1.98 (s, 3 H), 1.52 (d, *J* = 7.3 Hz, 3 H), 1.16 (t, *J* = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 172.2, 146.2, 139.1, 133.3, 130.1, 128.8, 127.6, 125.2, 61.0, 38.4, 21.2, 16.0, 14.0, 10.0 ppm. IR (film): $\tilde{\nu}$ = 2982, 2937, 1737, 1517, 1451, 1420, 1222, 1182, 1154, 1083, 1026, 826 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{16}H_{20}N_2O_2$ (M^+) 272.1525; found 272.1528.

Ethyl 2-[1-(4-Chlorophenyl)-5-methyl-1*H*-imidazol-2-yl]propanoate (5b): Pale yellow oil (208 mg, 0.71 mmol, 71%). ^1H NMR (400 MHz, $CDCl_3$): δ = 7.49 (dd, *J* = 7.6, 1.3 Hz, 2 H), 7.23–7.14 (br., 2 H), 6.85 (d, *J* = 0.8 Hz, 1 H), 4.12–3.98 (m, 2 H), 3.56 (q, *J* = 7.2 Hz, 1 H), 1.99 (d, *J* = 0.8 Hz, 3 H), 1.54 (d, *J* = 7.2 Hz, 3 H), 1.17 (t, *J* = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 171.9, 145.9, 135.0, 134.4, 129.7, 129.2, 128.6, 125.5, 61.0, 38.3, 15.8, 13.9, 9.9 ppm. IR (film): $\tilde{\nu}$ = 2983, 2938, 1735, 1494, 1450, 1419, 1376, 1314, 1221, 1182, 1091, 1012, 841 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{15}H_{17}N_2O_2Cl$ (M^+) 292.0979; found 292.0985.

Ethyl 2-[1-(3-Chlorophenyl)-5-methyl-1*H*-imidazol-2-yl]propanoate (5c): Pale yellow oil (165 mg, 0.57 mmol, 57%). ^1H NMR (400 MHz, $CDCl_3$): δ = 7.51–7.43 (m, 2 H), 7.27 (br., 1 H), 7.16 (br., 1 H), 6.85 (d, *J* = 0.8 Hz, 1 H), 4.14–3.98 (m, 2 H), 3.58 (q, *J* = 7.2 Hz, 1 H), 2.01 (d, *J* = 0.8 Hz, 3 H), 1.55 (d, *J* = 7.3 Hz, 3 H), 1.18 (t, *J* = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 171.9, 145.9, 137.1, 135.0, 130.5, 129.3, 128.6, 128.3, 126.2, 125.5, 61.1, 38.3, 15.8, 14.0, 9.9 ppm. IR (film): $\tilde{\nu}$ = 2984, 2938, 1735, 1594, 1484, 1222, 1182, 1077, 1026, 793, 693 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{15}H_{17}N_2O_2Cl$ (M^+) 292.0979; found 292.0972.

Ethyl 2-[1-(2-Chlorophenyl)-5-methyl-1*H*-imidazol-2-yl]propanoate (5d): Pale yellow oil (116 mg, 0.40 mmol, 40%). ^1H NMR (400 MHz, $CDCl_3$): δ = 7.58 (dd, *J* = 7.9, 1.6 Hz, 1 H), 7.48–7.46 (m, 1 H), 7.45–7.40 (m, 1 H), 7.35 (dd, *J* = 7.7, 1.8 Hz, 1 H), 6.89 (d, *J* = 1.0 Hz, 1 H), 4.09–4.00 (m, 2 H), 3.38 (q, *J* = 7.2 Hz, 1 H), 1.95 (d, *J* = 1.0 Hz, 3 H), 1.59 (d, *J* = 7.2 Hz, 3 H), 1.16 (t, *J* = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 Hz, $CDCl_3$): δ = 171.9, 145.8, 133.6, 133.0, 130.74, 130.69, 130.4, 128.5, 127.7, 125.6, 61.0, 38.4, 15.6, 14.0, 9.6 ppm. IR (film): $\tilde{\nu}$ = 2984, 2939, 1738, 1489, 1444, 1413, 1222, 1182, 1154, 1083, 1030, 770 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{15}H_{17}N_2O_2Cl$ (M^+) 292.0979; found 292.0975.

Ethyl 2-(1-Cyclohexyl-5-methyl-1*H*-imidazol-2-yl)propanoate (5e): Yellow oil (219 mg, 0.83 mmol, 83%). ^1H NMR (400 MHz, $CDCl_3$): δ = 6.70 (s, 1 H), 4.22–4.08 (m, 2 H), 4.04–3.89 (m, 2 H), 2.30 (s, 3 H), 1.97–1.80 (m, 6 H), 1.78–1.71 (m, 1 H), 1.63 (d, *J* = 7.2 Hz, 3 H), 1.45–1.29 (m, 2 H), 1.28–1.15 (m, 4 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 172.5, 145.4, 127.4, 127.0, 61.2, 56.3, 39.2, 32.1, 31.9, 26.30, 26.29, 25.3, 16.2, 14.1, 12.2 ppm. IR (film): $\tilde{\nu}$ = 2935, 2856, 1735, 1449, 1403, 1265, 1204, 1178, 1157, 1088, 1028 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{15}H_{24}N_2O_2$ (M^+) 264.1838; found 264.1841.

Ethyl 2-(1-Isopropyl-5-methyl-1*H*-imidazol-2-yl)propanoate (5f): Colorless oil (208 mg, 0.93 mmol, 93%). ^1H NMR (400 MHz, $CDCl_3$): δ = 6.70 (s, 1 H), 4.47 (hept, *J* = 7.0 Hz, 1 H), 4.23–4.09 (m, 2 H), 3.91 (q, *J* = 7.2 Hz, 1 H), 2.30 (s, 3 H), 1.63 (d, *J* = 7.2 Hz, 3 H), 1.47 (d, *J* = 7.0 Hz, 6 H), 1.23 (t, *J* = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 171.9, 144.7, 126.8, 126.4, 60.7, 46.9, 38.5, 21.5, 21.2, 15.7, 13.7, 11.3 ppm. IR (film): $\tilde{\nu}$ = 2981, 2938, 1738, 1465, 1420, 1373, 1262, 1187, 1071, 1028 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{12}H_{20}N_2O_2$ (M^+) 224.1525; found 224.1532.

Ethyl 2-(1-Ethyl-5-methyl-1*H*-imidazol-2-yl)propanoate (5g): Colorless oil (202 mg, 0.96 mmol, 96%). ^1H NMR (400 MHz, $CDCl_3$): δ = 6.73 (d, *J* = 0.7 Hz, 1 H), 4.22–4.08 (m, 2 H), 3.99–3.78 (m, 3 H), 2.20 (d, *J* = 0.9 Hz, 3 H), 1.64 (d, *J* = 7.2 Hz, 3 H), 1.31–1.25 (t, *J* = 7.2 Hz, 3 H), 1.23 (t, *J* = 7.2 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 172.2, 144.6, 127.2, 125.5, 61.2, 38.4, 38.0, 15.9, 15.7, 14.1, 9.6 ppm. IR (film): $\tilde{\nu}$ = 2982, 2938, 1732, 1673, 1438, 1376, 1259, 1205, 1183, 1084, 1027 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{11}H_{18}N_2O_2$ (M^+) 210.1368; found 210.1371.

Ethyl 2-[1-(*tert*-Butyl)-5-methyl-1*H*-imidazol-2-yl]propanoate (5h): Colorless oil (128 mg, 0.54 mmol, 54%). ^1H NMR (400 MHz, $CDCl_3$): δ = 6.68 (d, *J* = 0.7 Hz, 1 H), 4.23–4.08 (m, 3 H), 2.37 (d, *J* = 0.8 Hz, 3 H), 1.70 (s, 9 H), 1.63 (d, *J* = 7.0 Hz, 3 H), 1.21 (t, *J* = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 172.7, 147.4, 128.4, 128.1, 61.0, 58.0, 41.6, 31.9, 17.8, 15.9, 14.1 ppm. IR (film): $\tilde{\nu}$ = 2980, 2937, 1739, 1463, 1390, 1249, 1225, 1178, 1085, 1027, 669 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{13}H_{22}N_2O_2$ (M^+) 238.1681; found 238.1674.

General Procedure for the Synthesis of Imidazoles 6: Condition A: To a solution of **4** (0.5 mmol), **2a** (0.5 mmol), and Et_3N (0.5 mmol) in THF (2 mL) was added I_2 (1 mmol). The mixture was stirred under an O_2 atmosphere at room temperature for 2 h. The reaction solution was then diluted with CH_2Cl_2 (10 mL), washed with saturated sodium thiosulfate (10 mL), water, and brine, dried with an-

hydrous Na_2SO_4, and evaporated in vacuo. The residue was subject to silica gel column chromatography with ethyl acetate/petroleum ether (1:2, v/v) as eluent to give pure **6**.

Ethyl 2-[5-Formyl-1-(*p*-tolyl)-1*H*-imidazol-2-yl]propanoate (6a): Yellow oil (102 mg, 0.36 mmol, 71%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.58 (s, 1 H), 7.88 (s, 1 H), 7.32 (d, J = 8.3 Hz, 2 H), 7.26–7.08 (br., 2 H), 4.16–4.02 (m, 2 H), 3.65 (q, J = 7.2 Hz, 1 H), 2.45 (s, 3 H), 1.56 (d, J = 7.2 Hz, 3 H), 1.18 (t, J = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 178.1, 170.8, 153.3, 140.5, 139.8, 133.0, 132.2, 130.0, 127.5, 126.5, 61.4, 37.6, 21.1, 15.6, 13.9 ppm. IR (film): $\tilde{\nu}$ = 2984, 2939, 1737, 1682, 1531, 1515, 1471, 1451, 1278, 1163, 1084, 1025, 833, 771, 546 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{16}H_{18}N_2O_3$ (M^+) 286.1317; found 286.1318.

Ethyl 2-[1-(4-Chlorophenyl)-5-formyl-1*H*-imidazol-2-yl]propanoate (6b): Pale orange oil (96 mg, 0.32 mmol, 63%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.63 (s, 1 H), 7.88 (s, 1 H), 7.50–7.49 (m, 2 H), 7.32–7.15 (br., 2 H), 4.16–4.02 (m, 2 H), 3.62 (q, J = 7.2 Hz, 1 H), 1.58 (d, J = 7.2 Hz, 3 H), 1.19 (t, J = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 177.9, 170.6, 153.4, 141.6, 135.7, 133.5, 132.8, 129.7, 129.2, 128.0, 61.6, 37.7, 15.6, 13.9 ppm. IR (film): $\tilde{\nu}$ = 2986, 1738, 1681, 1532, 1496, 1474, 1447, 1164, 1091, 1013, 843 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{15}H_{15}N_2O_3Cl$ (M^+) 306.0771; found 306.0763.

Ethyl 2-[1-(3-Chlorophenyl)-5-formyl-1*H*-imidazol-2-yl]propanoate (6c): Yellow oil (80 mg, 0.26 mmol, 52%). ^{1}H NMR (600 MHz, $CDCl_3$): δ = 9.64 (s, 1 H), 7.89 (s, 1 H), 7.53 (d, J = 8.2 Hz, 1 H), 7.46 (t, J = 8.0 Hz, 1 H), 7.35–7.28 (br., 1 H), 7.22–7.14 (br., 1 H), 4.20–4.39 (m, 2 H), 3.70–3.63 (m, 1 H), 1.61 (d, J = 7.2 Hz, 3 H), 1.20 (t, J = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 177.9, 170.6, 153.3, 141.4, 136.1, 135.0, 132.8, 130.4, 130.0, 128.3 and 127.1, 126.4 and 125.0, 61.7, 37.7, 15.6, 14.0 ppm. IR (film): $\tilde{\nu}$ = 2986, 1738, 1681, 1593, 1533, 1480, 1445, 1163, 1077, 814, 687 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{15}H_{15}N_2O_3Cl$ (M^+) 306.0771; found 306.0780.

Ethyl 2-[1-(2-Chlorophenyl)-5-formyl-1*H*-imidazol-2-yl]propanoate (6d): Pale yellow oil (46 mg, 0.15 mmol, 30%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.64 (s, 1 H), 7.91 (s, 1 H), 7.58 (dd, J = 8.0, 1.2 Hz, 1 H), 7.52–7.48 (m, 1 H), 7.43–7.38 (m, 2 H), 4.16–4.03 (m, 2 H), 3.45 (q, J = 7.2 Hz, 1 H), 1.62 (d, J = 7.2 Hz, 3 H), 1.19 (t, J = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 177.9, 170.7, 153.3, 141.6, 133.2, 132.5, 132.1, 131.2, 130.4, 129.8, 127.8, 61.6, 37.9, 15.4, 14.0 ppm. IR (film): $\tilde{\nu}$ = 2923, 2850, 1737, 1679, 1536, 1489, 1469, 1377, 1162, 1086, 1026, 813 cm^{-1}. MS (MALDI-TOF): *m*/*z* = 307.10 [M + H]$^+$. HRMS (EI): *m*/*z* calcd. for $C_{15}H_{15}N_2O_3$ ([M – Cl]$^+$) 271.1083; found 271.1079.

Ethyl 2-(5-Formyl-1-phenyl-1*H*-imidazol-2-yl)propanoate (6e): Yellow oil (101 mg, 0.37 mmol, 74%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.59 (s, 1 H), 7.89 (s, 1 H), 7.57–7.49 (m, 3 H), 7.37–7.23 (br., 2 H), 4.15–4.03 (m, 2 H), 3.64 (q, J = 7.2 Hz, 1 H), 1.58 (d, J = 7.2 Hz, 3 H), 1.18 (t, J = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 178.2, 170.8, 153.3, 140.8, 134.9, 133.0, 129.8, 129.5, 128.0 and 126.8, 61.5, 37.8, 15.7, 14.0 ppm. IR (film): $\tilde{\nu}$ = 2985, 2940, 1737, 1682, 1597, 1532, 1498, 1473, 1450, 1278, 1163, 1076, 1026, 815, 776, 695 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{15}H_{16}N_2O_3$ (M^+) 272.1161; found 272.1163.

Ethyl 2-[5-Formyl-1-(4-methoxyphenyl)-1*H*-imidazol-2-yl]propanoate (6f): Yellow oil (106 mg, 0.35 mmol, 70%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.58 (s, 1 H), 7.87 (s, 1 H), 7.28–7.13 (br., 2 H), 7.01 (dd, J = 7.6, 1.5 Hz, 2 H), 4.17–4.02 (m, 2 H), 3.88 (s, 3 H), 3.66 (q, J = 7.2 Hz, 1 H), 1.57 (d, J = 7.2 Hz, 3 H), 1.19 (t, J = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 178.3, 170.9, 160.3, 153.6, 140.7, 133.1, 128.9 and 127.9, 127.3, 114.6, 61.5, 55.5, 37.7, 15.7, 14.0 ppm. IR (film): $\tilde{\nu}$ = 2984, 2939, 2839, 1737, 1682, 1531, 1514, 1470, 1454, 1300, 1252, 1164, 1024, 841 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{16}H_{18}N_2O_4$ (M^+) 302.1267; found 302.1270.

Ethyl 2-{5-Formyl-1-[4-(trifluoromethyl)phenyl]-1*H*-imidazol-2-yl}-propanoate (6g): Yellow oil (136 mg, 0.2 mmol, 40%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.65 (s, 1 H), 7.90 (s, 1 H), 7.80 (d, J = 8.6 Hz, 2 H), 7.50–7.37 (br., 2 H), 4.16–4.01 (m, 2 H), 3.60 (q, J = 7.2 Hz, 1 H), 1.59 (d, J = 7.2 Hz, 3 H), 1.18 (t, J = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 177.8, 170.5, 153.4, 142.0, 138.4, 132.8, 131.8 (q, J = 33.1 Hz), 129.6–127.8 (br.), 126.6 (q, J = 3.6 Hz), 123.4 (q, J = 271.7 Hz), 61.7, 37.8, 15.6, 13.9 ppm. IR (film): $\tilde{\nu}$ = 2988, 1738, 1682, 1616, 1533, 1441, 1326, 1166, 1129, 1068, 854, 605 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{16}H_{15}N_2O_3F_3$ (M^+) 340.1035; found 340.1036.

2-Benzhydryl-1-(*p*-tolyl)-1*H*-imidazole-5-carbaldehyde (6h): Yellow oil (121 mg, 0.35 mmol, 69%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.55 (s, 1 H), 7.92 (s, 1 H), 7.31–7.19 (m, 8 H), 7.16 (d, J = 7.2 Hz, 4 H), 6.97 (d, J = 8.0 Hz, 2 H), 5.19 (s, 1 H), 2.43 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 178.3, 155.9, 141.0, 140.2, 139.7, 132.7, 132.5, 130.0, 128.8, 128.7, 128.5, 127.1, 48.3, 21.3 ppm. IR (film): $\tilde{\nu}$ = 3027, 1682, 1531, 1514, 1494, 1454, 1434, 1157, 821, 725, 701, 542 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{24}H_{20}N_2O$ (M^+) 352.1576; found 352.1577.

2-Benzhydryl-1-(4-methoxyphenyl)-1*H*-imidazole-5-carbaldehyde (6i): Yellow oil (138 mg, 0.38 mmol, 75%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.57 (s, 1 H), 7.92 (s, 1 H), 7.31–7.20 (m, 6 H), 7.18–7.13 (m, 4 H), 7.00–6.98 (m, 2 H), 6.95–6.91 (m, 2 H), 5.20 (s, 1 H), 3.87 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 178.4, 160.2, 156.1, 141.1, 140.3, 132.8, 128.8, 128.6, 128.5, 127.6, 127.1, 114.5, 55.5, 48.4 ppm. IR (film): $\tilde{\nu}$ = 3025, 1678, 1530, 1513, 1454, 1251, 1157, 1025, 837, 726, 701 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{24}H_{20}N_2O_2$ (M^+) 368.1525; found 368.1526.

2-Benzhydryl-1-(4-bromophenyl)-1*H*-imidazole-5-carbaldehyde (6j): Yellow oil (135 mg, 0.33 mmol, 65%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.60 (s, 1 H), 7.91 (s, 1 H), 7.57–7.54 (m, 2 H), 7.32–7.20 (m, 6 H), 7.17–7.11 (m, 4 H), 6.95–6.91 (m, 2 H), 5.15 (s, 1 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 178.0, 156.0, 141.9, 139.9, 134.4, 132.6, 132.5, 128.9, 128.69, 128.66, 127.3, 123.8, 48.5 ppm. IR (film): $\tilde{\nu}$ = 3061, 1678, 1531, 1493, 1452, 1433, 1158, 1068, 1006, 834, 732, 702, 540 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{23}H_{17}N_2OBr$ (M^+) 416.0524; found 416.0527.

Ethyl 2-[5-Formyl-1-(*p*-tolyl)-1*H*-imidazol-2-yl]-2-phenylacetate (6k): Yellow oil (96 mg, 0.28 mmol, 55%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.56 (s, 1 H), 7.90 (s, 1 H), 7.38–7.11 (m, 8 H), 6.85 (br., 1 H), 4.80 (s, 1 H), 4.19–4.13 (m, 2 H), 2.45 (s, 3 H), 1.19 (t, J = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 178.3, 169.0, 151.6, 140.6, 140.0, 134.4, 133.0, 132.1, 130.1, 129.2, 128.5, 128.0, 127.1, 61.9, 49.5, 21.3, 14.0 ppm. IR (film): $\tilde{\nu}$ = 2981, 1747, 1682, 1530, 1514, 1454, 1191, 1159, 1029, 829, 697, 541 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{21}H_{20}N_2O_3$ (M^+) 348.1474; found 348.1473.

Ethyl 2-(4-Bromophenyl)-2-[5-formyl-1-(*p*-tolyl)-1*H*-imidazol-2-yl]-acetate (6l): Brown solid (98 mg, 0.23 mmol, 46%), m.p. 83–87 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.48 (s, 1 H), 7.81 (s, 1 H), 7.37–7.33 (m, 2 H), 7.33–7.28 (m, 2 H), 7.18–7.09 (br., 2 H), 7.06–6.98 (br., 1 H), 6.87–6.78 (br., 1 H), 4.38 (s, 1 H), 4.27–4.14 (m, 2 H), 2.37 (s, 3 H), 1.25 (t, J = 7.1 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 178.9, 171.1, 152.8, 139.6, 137.7, 136.7, 134.3, 131.8, 130.8, 129.1, 128.5, 128.0, 122.6, 76.5, 63.9, 21.2, 13.7 ppm. IR

(film): $\tilde{\nu}$ = 2982, 1737, 1682, 1529, 1514, 1486, 1444, 1247, 1159, 1086, 1010, 828, 770, 732, 539 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{21}H_{19}N_2O_3Br$ (M^+) 426.0579; found 426.0582.

Procedure for the Synthesis of 2-Benzhydryl-1-(*p*-tolyl)-1,8-dihydroindeno[1,2-*d*]imidazole (7): To a 25 mL pressure-tube charged with a magnetic stir bar was added **3s** (99 mg, 0.2 mmol), $Pd(OAc)_2$ (3 mg, 0.01 mmol), PPh_3 (6 mg, 0.02 mmol), Cs_2CO_3 (130 mg, 0.4 mmol), $(n\text{-}Bu)_4NBr$ (13 mg, 0.04 mmol), and CH_3CN (2 mL) under a N_2 atmosphere. The reaction vessel was sealed and heated to 150 °C in an oil bath for 10 h. The reaction mixture was cooled to room temperature and then the solvent was evaporated in vacuo. The residue was subject to silica gel column chromatography with ethyl acetate/petroleum ether (1:5, v/v) as eluent to give **7** as a pale yellow solid (45 mg, 0.11 mmol, 55%), m.p. 203–205 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 7.71 (d, J = 7.5 Hz, 1 H), 7.35 (d, J = 7.4 Hz, 1 H), 7.32–7.23 (m, 11 H), 7.23–7.17 (m, 2 H), 7.11–7.07 (m, 3 H), 5.40 (s, 1 H), 3.45 (s, 2 H), 2.43 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 152.4, 146.6, 142.8, 141.7, 138.8, 138.0, 137.8, 134.4, 130.2, 129.0, 128.4, 126.9, 126.6, 126.0, 125.0, 123.6, 118.3, 49.1, 28.7, 21.2 ppm. IR (film): $\tilde{\nu}$ = 3058, 3026, 1611, 1515, 1493, 1456, 1397, 1167, 1007, 909, 763, 726, 700 cm^{-1}. HRMS (EI): *m*/*z* calcd. for $C_{30}H_{24}N_2$ (M^+) 412.1939; found 412.1943.

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[13] For ORTEP plots of product **3d**, please see the Supporting Information. CCDC-1000726 contains supplementary crystallographic data for compound **3d**. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.

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