Indium(III) Chloride Catalyzed One-Pot Multicomponent Synthesis of Chromenone- and Quinolone-Annulated Imidazole Derivatives

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Abstract: A one-pot multicomponent strategy for the synthesis of chromenone- and quinolone-annulated imidazole derivatives from easily available starting materials has been achieved via an indium(III) chloride catalyzed domino reaction. The protocol is simple, step-economic, and less hazardous, and also provides good yields of the products.

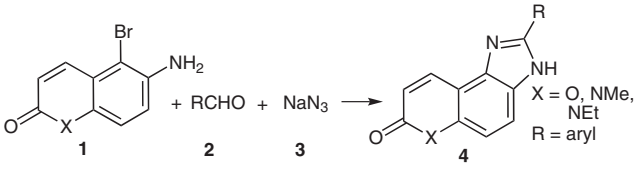
Key words: multicomponent reaction, indium(III) chloride, imidazole, aromatic aldehyde, sodium azide

Among various five-membered nitrogen heterocycles, imidazole derivatives have gained a distinctive place in the field of medicinal chemistry. The insertion of the imidazole nucleus is a vital synthetic strategy in drug design for its well-known biological properties, which includes antibacterial,[1] antifungal,[2] analgesic,[3] anti-inflammatory,[4] anticancer,[5] antiviral,[6] and antidepressant[7] properties. Furthermore, some compounds bearing a chromenone-annulated imidazole core are effective as a phosphodiesterase inhibitor,[8] CNS depressant,[9] and also inhibit carcinoma in mammals.[10] Many compounds of industrial and technological importance contain the imidazole nucleus, and it can also be found in various compounds that are used in photography and electronics.[11]

Furthermore, chromenones and quinolone subunits are the parent component of a large number of bioactive natural products, and their derivatives show extensive biological activity.[12] Chromeno[5,6-*d*]imidazol-7(3*H*)-one and 3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one cores posses an imidazole nucleus fused to a chromenone and quinolone molecule respectively at their 5,6-position. This new class of compounds and their derivatives may be of interest to pharmacologists and medicinal chemists. Close analogues, chromeno[4,3-*d*]imidazol-4(3*H*)-one derivatives, were synthesized by Trimarco et al.[13] via a multistep synthesis, but chromeno[5,6-*d*]imidazol-7(3*H*)-one and 3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one are still less investigated and to the best of our knowledge so far there is only one report[14] of chromeno[5,6-*d*]imidazol-7(3*H*)-one derivatives by the reaction of 5,6-diamino-2*H*-chromen-2-one, benzoic acid, and polyphosphoric acid under reflux conditions; the methodology suffers from the requirement for strongly acidic conditions. Hence the synthesis of chromeno[5,6-*d*]imidazol-7(3*H*)-one and 3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one derivatives by a simple, step-economic, and less hazardous methodology is still important and requires further investigation.

Multicomponent reactions (MCR) play an important role in synthetic and medicinal chemistry and have been utilized to access bioactive compounds.[15] Their ability to generate complex molecular scaffolds from simple precursors in a single step makes them a powerful synthetic tool for organic synthesis. Multicomponent reactions are advantageous compared to linear stepwise synthesis as they reduce reaction times and provide high atom economy and selectivity.[16]

Recently Lee et al.[17] reported a copper-catalyzed three-component reaction of a 2-haloaniline, an aldehyde, and sodium azide for the construction of a fused imidazole ring. Our interest in the synthesis of potential bioactive heterocycles by metal catalysis and multicomponent strategy[18] prompted us to undertake a study of the synthesis of chromeno[5,6-*d*]imidazol-7(3*H*)-one, and 3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one derivatives by the metal-catalyzed multicomponent reaction approach; herein, we report our results (Scheme 1).



Scheme 1 *Reagents and conditions*: $InCl_3$ (10 mol%), DMSO, 130 °C, 8 h.

As a model reaction, 6-amino-5-bromo-2*H*-chromen-2-one (**1a**), 4-methoxybenzaldehyde (**2a**), and sodium azide (**3**) were reacted in dimethyl sulfoxide using copper(I) iodide (10 mol%) as the catalyst at 130 °C for eight hours. A new product was obtained in low yield (47%) (Table 1, entry 1). The spectral analysis of the compound confirmed the formation of the desired product **4a**. As this type of reaction may be improved by introduction of a ligand, we examined the use of ligands such as *N*,*N*,*N*′,*N*′-tetramethylethylenediamine, *N*,*N*′-dimethylethylenediamine, and L-proline in this reaction, however their use did not improve the yield of product **4a** (entries 2–4). Copper(II) was also inefficient in enhancing the yield of the reaction (entry 5). We next considered the use of indium(III) chloride as it is known for its relatively low toxicity and excellent tolerance to oxygen- and nitrogen-containing

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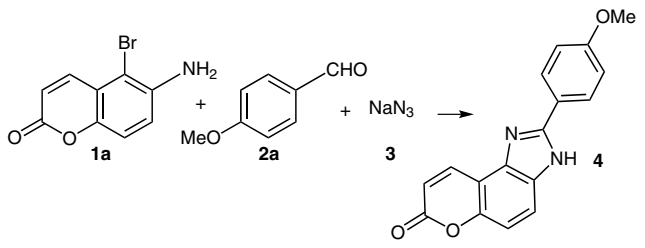
Table 1 Optimization of the Reaction Conditions[a]



Entry	Catalyst (mol%)	Solvent	Temp (°C)	Yield[b] (%)
1	CuI (10)	DMSO	130	47
2[c]	CuI (10)	DMSO	130	49
3[d]	CuI (10)	DMSO	130	45
4[e]	CuI (10)	DMSO	130	39
5	$Cu(OAc)_2$ (10)	DMSO	130	36
6[f]	$InCl_3$ (10)	DMSO	130	78
7	$InCl_3$ (10)	DMSO	150	75
8	$InCl_3$ (10)	DMSO	120	52
9	$InCl_3$ (5)	DMSO	130	72
10	$InCl_3$ (20)	DMSO	130	69
11	$InCl_3$ (10)	DMF	130	53
12	$InCl_3$ (10)	xylene	130	0
13	$InCl_3$ (10)	toluene	reflux	0
14	$Yb(OTf)_2$ (10)	DMSO	130	trace
15	$AlCl_3$ (10)	DMSO	130	0
16	$FeCl_3$ (10)	DMSO	130C	0
17	$AgSbF_6$ (10)	DMSO	130	0
18	–	DMSO	130	0

[a] Reaction conditions: 6-amino-5-bromo-2*H*-chromen-2-one (**1a**, 1.0 mmol), 4-methoxybenzaldehyde (**2a**, 1.5 mmol), NaN_3 (**3**, 2.0 mmol), catalyst, solvent (5 mL), 8 h.
[b] Isolated yields.
[c] TMEDA (10 mol%) was used as a ligand.
[d] DMEDA (10 mol%) was used as a ligand.
[e] L-proline (10 mol%) was used as a ligand.
[f] Optimized reaction condition.

substrates and functional groups.[19] When indium(III) chloride was employed as the catalyst in dimethyl sulfoxide at 130 °C, a satisfactory result was obtained without the use of a ligand (78%; entry 6). The yield of the product was not improved by increasing or decreasing the reaction temperature (entries 7 and 8). We also found that the yield of the reaction also did not improved by increasing or decreasing the catalyst loading (entries 9 and 10). We also examined the used of different solvents, such as *N*,*N*-dimethylformamide, xylene, and toluene; *N*,*N*-dimethylformamide was moderately effective (entry 11) but nonpolar xylene and toluene were ineffective (entries 12 and 13). Other Lewis acid catalysts [$Yb(OTf)_2$, $AlCl_3$, $FeCl_3$, $AgSbF_6$] were also found to be ineffective (entries 14–17). This reaction failed completely without a catalyst (entry 18).

Thus, the optimized reaction conditions are 10 mol% indium(III) chloride in dimethyl sulfoxide at 130 °C for eight hours. The scope of this protocol was extended to different aromatic and heteroaromatic aldehydes and other heterocyclic systems.

The results showed that the precursors **1a**–**c** successfully gave the desired chromeno[5,6-*d*]imidazol-7(3*H*)-one and 3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one derivatives in very good yields (68–82%). The structures of the products **4a**–**m** were determined from their spectral and analytical data. ^{1}H NMR of the product **4a** showed a characteristic peak for the NH proton at δ = 13.26 as well as at 13.21. Moreover, it also showed two pairs of doublets, one pair at δ = 6.61 (*J* = 9.6 Hz) and 6.53 (*J* = 9.6 Hz), and another at 7.84 (*J* = 8.8Hz) and 7.72 (*J* = 8.4Hz). The pair of singlets and both the pairs of doublets and also their coupling constants indicate that the compound **4a** might exist in two tautomeric forms **4a** and **4a′** (Figure 1). These structures were fully optimized by the Gaussian 03 program using the DFT method (using B3LYP level and 6-31G as basis set). The calculations show that the tautomer **4a** (–621199.57 kcal/mol) is more stable than the tautomer **4a′** (–621197.22 kcal/mol), thus it predominates.

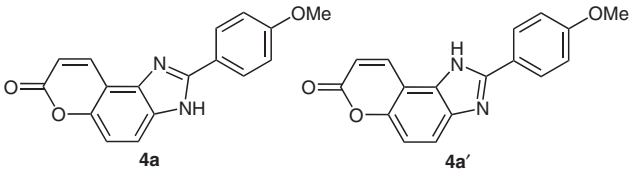


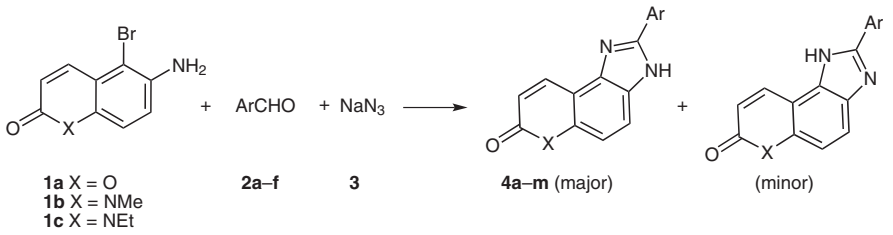
Figure 1 Two probable tautomers **4a** and **4a′**

The other compounds **4d**,**h**,**i**,**l**,**m** show both tautomers in their ^{1}H NMR spectra. The remaining compounds in Table 2 show only the major tautomer in their ^{1}H NMR spectra.

Aldehydes containing electron-donating groups furnished the corresponding imidazole derivatives in very good yields. Furfuraldehyde (**2e**) also gave the desired products in good yields. Aromatic aldehydes with electron-withdrawing groups, such as nitro or fluoro, yielded inseparable complex mixtures. The reaction was found to be totally ineffective for aliphatic aldehydes.

A plausible rationalization for the formation of the product **4** is depicted in Scheme 2. At first the condensation of the substrates **1** and aldehydes **2** generate an imine. The initially formed imine can follow either path a or path b. In path a, indium may co-ordinate with the imine bond **A** followed by azide insertion to give intermediate **C**. This may then undergo cyclization to form intermediate **E** that readily generates the product **4** by aromatization.[17] Alter-

Table 2 Synthesis of Various Chromeno[5,6-*d*]imidazol-7(3*H*)-one and 3,6-Dihydro-7*H*-Imidazo[4,5-*f*]quinolin-7-one Derivatives[a]

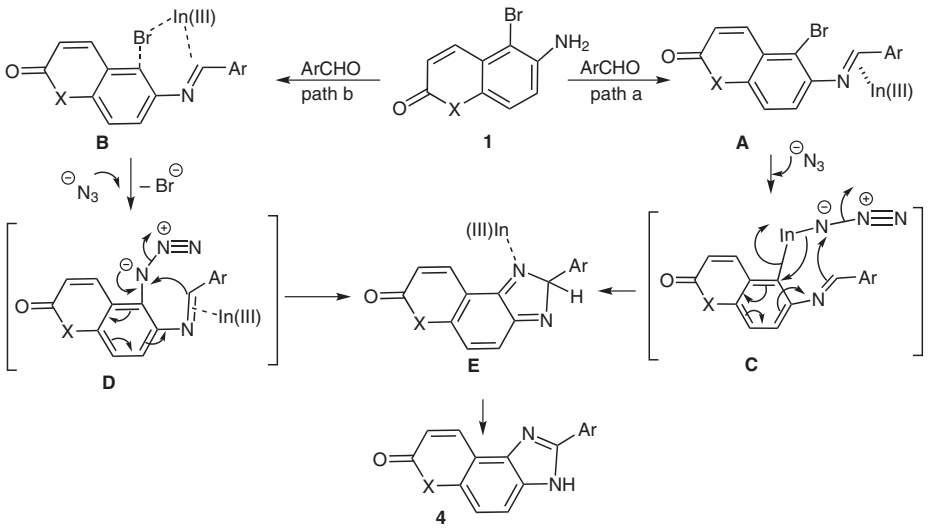


Entry	Precursor	Aldehyde	Product	Yield[b] (%)
1	**1a**	**2a**, Ar = 4-MeOC$_6$H$_4$	**4a**	78
2	**1a**	**2b**, Ar = Ph	**4b**	76
3	**1a**	**2c**, Ar = 4-MeC$_6$H$_4$	**4c**	74
4	**1a**	**2d**, Ar = 4-EtOC$_6$H$_4$	**4d**	81
5	**1a**	**2e**, Ar = 2-furyl	**4e**	75
6	**1a**	**2f**, Ar = 2-MeO-5-ClC$_6$H$_3$	**4f**	71
7	**1b**	**2b**, Ar = Ph	**4g**	70
8	**1b**	**2d**, Ar = 4-EtOC$_6$H$_4$	**4h**	74
9	**1c**	**2a**, Ar = 4-MeOC$_6$H$_4$	**4i**	80
10	**1c**	**2b**, Ar = Ph	**4j**	77
11	**1c**	**2c**, Ar = 4-MeC$_6$H$_4$	**4k**	73
12	**1c**	**2d**, Ar = 4-EtOC$_6$H$_4$	**4l**	82
13	**1c**	**2e**, Ar = 2-furyl	**4m**	74

[a] Reaction conditions: **1a**–**c** (1.0 mmol), aldehyde (1.5 mmol), NaN_3 (2.0 mmol), $InCl_3$ (0.1 mmol), DMSO (5 mL), 130 °C, 8 h.
[b] Isolated yield.

natively in path b, indium may coordinate with both the imine and the halogen **B** facilitating the replacement of halogen[20] by the azide forming the intermediate **D**. This may then undergo cyclization forming the common intermediates **E** that afford the products **4**.

In summary, we have developed an efficient protocol for the synthesis of chromeno[5,6-*d*]imidazol-7(3*H*)-one and 3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one derivatives in good yields. The advantages of this methodology are its simplicity and step economy, and it affords the products



Scheme 2 Plausible mechanistic pathway for the formation of **4**

in good yields. The synthesized compounds as well as the synthetic method could be useful to the pharmacologists and the medicinal chemists.

Melting ranges were determined in open capillaries and are uncorrected. IR spectra were recorded on a Perkin-Elmer L 120-000A spectrophotometer on KBr disks. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker DPX-400 spectrometer in DMSO-d_6 with TMS as internal standard. CHN was recorded on 2400 series II CHN analyzer Perkin Elmer. MS were recorded on a Q-TOF micro instrument at the Indian Institute of Chemical Biology, Kolkata. Silica gel [(60–120, 230–400 mesh), Rankem, India] was used for chromatographic separation. Silica gel G, GF-254 [CDH, (India)] was used for TLC. Petroleum ether refers to the fraction boiling between 60 °C and 80 °C.

2-(4-Methoxyphenyl)chromeno[5,6-*d*]imidazol-7(3*H*)-one (4a); Typical Procedure

6-Amino-5-bromo-2*H*-chromen-2-one (**1a**, 0.24 g, 1 mmol), 4-methoxybenzaldehyde (**2a**, 0.204 g, 1.5 mmol), and NaN_3 (**3**, 0.195 g, 2.0 mmol) were mixed in DMSO (5 mL). To the mixture was added $InCl_3$ (0.022 g, 0.1 mmol), and it was then heated with stirring at 130 °C for 8 h. After completion of the reaction (TLC monitoring), the mixture was cooled and diluted with EtOAc (50 mL), washed with H_2O (2 × 20 mL) and brine (20 mL), and dried (Na_2SO_4). The solvent was removed under reduced pressure. The resulting crude product was then purified by column chromatography (silica gel, 60–120 mesh, hexane–EtOAc, 1:1) to give **4a** (0.228 g, 0.78 mmol, 78%) as a light yellow solid; mp 264–266 °C.

IR (KBr): 3261, 1720, 1607, 1247, 772 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = [13.26 (br s), 13.21 (br s) (1 H)], 8.54 (d, *J* = 9.6 Hz, 1 H), 8.15 (d, *J* = 8.4 Hz, 2 H), [7.85 (d, *J* = 8.8 Hz), 7.72 (d, *J* = 8.4 Hz) (1 H)], 7.24 (d, *J* = 8.4 Hz, 1 H), 7.14 (d, *J* = 8.4 Hz, 2 H), [6.61 (d, *J* = 9.6 Hz), 6.53 (d, *J* = 9.6 Hz) (1 H)], 3.85 (s, 3 H).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 160.9, 160.6, 153.3, 150.0, 140.0, 139.6, 131.0, 128.2, 121.9, 115.1, 114.7, 114.4, 110.6, 109.3, 55.3.

HRMS: *m*/*z* [M + Na]$^+$ calcd for $C_{17}H_{12}N_2O_3$: 315.0746; found: 315.0749.

2-Phenylchromeno[5,6-*d*]imidazol-7(3*H*)-one (4b)

Following the typical procedure using **1a** (0.24 g) gave **4b** (0.20 g, 76%) as a pale yellow solid; mp >300 °C.

IR (KBr): 3245, 1683, 1605, 1287, 774 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 13.38 (br s, 1 H), 8.56 (d, *J* = 9.6 Hz, 1 H), 8.21 (d, *J* = 7.6 Hz, 2 H), 7.81 (br s, 1 H), 7.62–7.52 (m, 3 H), 7.28 (d, *J* = 8.8 Hz, 1 H), 6.58 (d, *J* = 8.8 Hz, 1 H).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 160.1, 150.3, 139.5, 131.7, 131.6, 130.3, 129.6, 129.1, 129.0, 126.6, 115.3, 111.2, 105.3, 99.5.

MS: *m*/*z* = 263.05 [M + H]$^+$.

Anal. Calcd for $C_{16}H_{10}N_2O_2$: C, 73.27; H, 3.84; N, 10.68. Found: C, 73.41; H, 3.88; N, 10.57.

2-*p*-Tolylchromeno[5,6-*d*]imidazol-7(3*H*)-one (4c)

Following the typical procedure using **1a** (0.24 g) gave **4c** (0.205 g, 74%) as a light yellow solid; mp 280–282 °C.

IR (KBr): 3456, 1727, 1608, 1290, 780 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 13.30 (br s, 1 H), 8.55 (d, *J* = 9.6 Hz, 1 H), 8.10 (d, *J* = 8.0 Hz, 2 H), 7.78 (br s, 1 H), 7.40 (d, *J* = 8.0 Hz, 2 H), 7.25 (d, *J* = 8.8 Hz, 1 H), 6.57 (d, *J* = 9.2 Hz, 1 H), 2.40 (s, 3 H).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 160.4, 150.3, 140.1, 139.6, 135.1, 131.6, 129.6, 128.7, 126.9, 126.5, 115.2, 110.9, 21.0.

MS: *m*/*z* = 277.03 [M + H]$^+$.

Anal. Calcd for $C_{17}H_{12}N_2O_2$: C, 73.90; H, 4.38; N, 10.14. Found: C, 74.09; H, 4.31; N, 10.08.

2-(4-Ethoxyphenyl)chromeno[5,6-*d*]imidazol-7(3*H*)-one (4d)

Following the typical procedure using **1a** (0.24 g) gave **4d** (0.248 g, 81%) as a pale yellow solid; mp 236–238 °C.

IR (KBr): 3221, 1706, 1608, 1255, 778 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = [13.23 (br s), 13.18 (br s) (1 H)], 8.53 (d, *J* = 9.2 Hz, 1 H), 8.14 (d, *J* = 8.4 Hz, 2 H), [7.85 (d, *J* = 8.4 Hz), 7.72 (d, *J* = 8.8 Hz) (1 H)], 7.23 (d, *J* = 8.4 Hz, 1 H), 7.12 (d, *J* = 8.4 Hz, 2 H), [6.61 (d, *J* = 9.6 Hz), 6.53 (d, *J* = 9.6 Hz) (1 H)], 4.12 (q, *J* = 6.8 Hz, 2 H), 1.37 (t, *J* = 6.8 Hz, 3 H).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 160.8, 160.1, 152.2, 140.9, 134.5, 133.7, 129.4, 128.7, 128.1, 120.2, 114.9, 111.4, 109.3, 63.4, 14.6.

MS: *m*/*z* = 307.13 [M + H]$^+$.

Anal. Calcd for $C_{18}H_{14}N_2O_3$: C, 70.58; H, 4.61; N, 9.15. Found: C, 70.74; H, 4.52; N, 9.03.

2-(Furan-2-yl)chromeno[5,6-*d*]imidazol-7(3*H*)-one (4e)

Following the typical procedure using **1a** (0.24 g) gave **4e** (0.189 g, 75%) as a brown solid; mp >300 °C.

IR (KBr): 3193, 1679, 1602, 1265, 780 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 13.42 (br s, 1 H), 8.51 (d, *J* = 9.2 Hz, 1 H), 7.98 (s, 1 H), 7.74 (br s, 1 H), 7.24 (d, *J* = 8.8 Hz, 2 H), 6.76 (s, 1 H), 6.54 (d, *J* = 9.2 Hz, 1 H).

MS: *m*/*z* = 253.07 [M + H]$^+$.

Anal. Calcd for $C_{14}H_8N_2O_3$: C, 66.67; H, 3.20; N, 11.11. Found: C, 66.43; H, 3.23; N, 11.17.

2-(5-Chloro-2-methoxyphenyl)chromeno[5,6-*d*]imidazol-7(3*H*)-one (4f)

Following the typical procedure using **1a** (0.24 g) gave **4f** (0.232 g, 71%) as a light reddish solid; mp >300 °C.

IR (KBr): 32345, 1714, 1604, 1278, 772 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.60 (br s, 1 H), 8.59 (d, *J* = 9.6 Hz, 1 H), 8.38 (d, *J* = 2.8 Hz, 1 H), 7.85 (d, *J* = 8.8 Hz, 1 H), 7.57 (dd, *J* = 8.8, 2.8 Hz, 1 H), 7.31 (t, *J* = 8.8 Hz, 2 H), 6.55 (d, *J* = 9.6 Hz, 1 H), 4.06 (s, 3 H).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 160.6, 155.6, 150.2, 149.2, 139.6, 138.8, 131.1, 131.0, 128.7, 124.8, 116.0, 115.3, 114.4, 111.4, 109.3, 56.4.

MS: *m*/*z* = 327.05 [M + H]$^+$, 329.06 [M + H + 2]$^+$.

Anal. Calcd for $C_{17}H_{11}ClN_2O_3$: C, 62.49; H, 3.39; N, 8.57. Found: C, 62.22; H, 3.44; N, 8.69.

6-Methyl-2-phenyl-3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one (4g)

Following the typical procedure using **1b** (0.253 g) gave **4g** (0.193 g, 70%) as a light yellow solid; mp >300 °C.

IR (KBr): 3113, 1636, 1595, 1287, 779 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 13.23 (br s, 1 H), 8.51 (d, *J* = 9.6 Hz, 1 H), 8.22 (d, *J* = 7.6 Hz, 2 H), 7.82 (br s, 1 H), 7.59 (t, *J* = 7.2 Hz, 2 H), 7.52 (t, *J* = 7.2 Hz, 1 H), 7.43 (d, *J* = 9.2 Hz, 1 H), 6.75 (br s, 1 H), 3.72 (s, 3 H).

MS: *m*/*z* = 276.06 [M + H]$^+$.

Anal. Calcd for $C_{17}H_{13}N_3O$: C, 74.17; H, 4.76; N, 15.26. Found: C, 74.28; H, 4.80; N, 15.17.

2-(4-Ethoxyphenyl)-6-methyl-3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one (4h)
Following the typical procedure using **1b** (0.253 g) gave **4h** (0.236 g, 74%) as a pale yellow solid; mp >300 °C.

IR (KBr): 3224, 1645, 1597, 1247, 778 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = [13.08 (br s), 13.04 (br s) (1 H)], 8.47 (d, *J* = 9.2 Hz, 1 H), 8.13 (d, *J* = 7.6 Hz, 2 H), [7.87 (d, *J* = 8.8 Hz), 7.74 (d, *J* = 8.8 Hz) (1 H)], 7.38 (d, *J* = 8.8 Hz, 1 H), 7.11–7.09 (m, 2 H), [6.74 (d, *J* = 9.2 Hz), 6.68 (d, *J* = 9.2 Hz) (1 H)], 4.11 (q, *J* = 6.8 Hz, 2 H), 3.69 (s, 3 H), 1.35 (t, *J* = 6.8 Hz, 3 H).

MS: *m*/*z* = 320.14 $[M + H]^+$.

Anal. Calcd for $C_{19}H_{17}N_3O_2$: C, 71.46; H, 5.37; N, 13.16. Found: C, 71.28; H, 5.40; N, 13.27.

6-Ethyl-2-(4-methoxyphenyl)-3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one (4i)
Following the typical procedure using **1c** (0.267 g) gave **4i** (0.255 g, 80%) as a light brown solid; mp >300 °C.

IR (KBr): 3084, 1643, 1601, 1256, 786 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = [13.07 (br s), 13.03 (br s) (1 H)], 8.49 (d, *J* = 9.6 Hz, 1 H), 8.18–8.14 (m, 2 H), [7.88 (d, *J* = 9.2 Hz), 7.76 (d, *J* = 9.2 Hz) (1 H)], 7.43 (dd, *J* = 8.8, 4.4 Hz, 1 H), 7.14 (dd, *J* = 8.0, 4.0 Hz, 2 H), [6.74 (d, *J* = 9.6 Hz), 6.68 (d, *J* = 9.6 Hz) (1 H)], 4.37 (q, *J* = 6.8 Hz, 2 H), 3.86 (s, 3 H), 1.27 (t, *J* = 6.8 Hz, 3 H).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 160.7, 160.5, 152.2, 140.8, 134.8, 133.6, 129.3, 128.1, 128.0, 122.4, 120.2, 120.0, 114.4, 111.3, 109.3, 55.4, 37.4, 12.9.

MS: *m*/*z* = 320.05 $[M + H]^+$.

Anal. Calcd for $C_{19}H_{17}N_3O_2$: C, 71.46; H, 5.37; N, 13.16. Found: C, 71.59; H, 5.31; N, 13.12.

6-Ethyl-2-phenyl-3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one (4j)
Following the typical procedure using **1c** (0.267 g) gave **4j** (0.233 g, 77%) as a light yellow solid; mp >300 °C.

IR (KBr): 3109, 1648, 1598, 1250, 782 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 13.23 (br s, 1 H), 8.51 (d, *J* = 9.6 Hz, 1 H), 8.22 (d, *J* = 8.0 Hz, 2 H), 7.90 (br s, 1 H), 7.59 (t, *J* = 7.2 Hz, 2 H), 7.53 (d, *J* = 7.2 Hz, 1 H), 7.48 (d, *J* = 9.2 Hz, 1 H), 6.73 (d, *J* = 8.0 Hz, 1 H), 4.38 (q, *J* = 6.8 Hz, 2 H), 1.27 (t, *J* = 6.8 Hz, 3 H).

MS: *m*/*z* = 290.10 $[M + H]^+$.

Anal. Calcd for $C_{18}H_{15}N_3O$: C, 74.72; H, 5.23; N, 14.52. Found: C, 74.94; H, 5.19; N, 14.44.

6-Ethyl-2-*p*-tolyl-3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one (4k)
Following the typical procedure using **1c** (0.267 g) gave **4k** (0.221 g, 73%) as a yellow solid; mp >300 °C.

IR (KBr): 3115, 1644, 1599, 1285, 787 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 13.14 (br s, 1 H), 8.50 (d, *J* = 8.4 Hz, 1 H), 8.11 (d, *J* = 7.6 Hz, 2 H), 7.78 (d, *J* = 8.0 Hz, 1 H), 7.46 (d, *J* = 8.0 Hz, 1 H), 7.39 (d, *J* = 7.6 Hz, 2 H), 6.69 (d, *J* = 9.2 Hz, 1 H), 4.37 (q, *J* = 6.8 Hz, 2 H), 2.40 (s, 3 H), 1.27 (t, *J* = 6.8 Hz, 3 H).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 161.2, 152.7, 141.3, 140.3, 135.4, 134.2, 130.1, 129.8, 127.0, 126.9, 120.9, 120.6, 115.1, 111.9, 110.2, 109.9, 37.8, 21.5, 13.5.

MS: *m*/*z* = 304.06 $[M + H]^+$.

Anal. Calcd for $C_{19}H_{17}N_3O$: C, 75.23; H, 5.65; N, 13.85. Found: C, 75.39; H, 5.62; N, 13.79.

2-(4-Ethoxyphenyl)-6-ethyl-3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one (4l)
Following the typical procedure using **1c** (0.267 g) gave **4l** (0.273 g, 82%) as a light yellow solid; mp 288–290 °C.

IR (KBr): 3123, 1643, 1602, 1260, 786 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = [13.07 (br s), 13.03 (br s) (1 H)], 8.48 (d, *J* = 9.6 Hz, 1 H), 8.14 (d, *J* = 7.6 Hz, 2 H), [7.88 (d, *J* = 8.4 Hz), 7.75 (d, *J* = 8.8 Hz) (1 H)], 7.43 (d, *J* = 9.2 Hz, 1 H), 7.12 (d, *J* = 7.6 Hz, 2 H), [6.74 (d, *J* = 9.2 Hz), 6.68 (d, *J* = 9.2 Hz) (1 H)], 4.36 (q, *J* = 6.4 Hz, 2 H), 4.13 (q, *J* = 6.8 Hz, 2 H), 1.37 (t, *J* = 6.8 Hz, 3 H), 1.26 (t, *J* = 6.4 Hz, 3 H).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 161.2, 160.5, 152.7, 141.4, 139.0, 135.3, 134.2, 129.8, 128.6, 122.7, 122.4, 120.7, 120.5, 115.4, 111.8, 109.8, 63.8, 37.7, 15.1, 13.4.

MS: *m*/*z* = 334.09 $[M + H]^+$.

Anal. Calcd for $C_{20}H_{19}N_3O_2$: C, 72.05; H, 5.74; N, 12.60. Found: C, 71.92; H, 5.81; N, 12.69.

6-Ethyl-2-(furan-2-yl)-3,6-dihydro-7*H*-imidazo[4,5-*f*]quinolin-7-one (4m)
Following the typical procedure using **1c** (0.267 g) gave **4m** (0.207 g, 74%) as a deep yellow solid; mp 290–292 °C.

IR (KBr): 3117, 1647, 1598, 1236, 785 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = [13.44 (br s), 13.21 (br s) (1 H)], [8.48 (d, *J* = 9.6 Hz), 8.42 (d, *J* = 9.6 Hz) (1 H)], 7.95 (br s, 1 H), [7.85 (d, *J* = 9.2 Hz), 7.73 (d, *J* = 8.8 Hz) (1 H)], 7.45 (t, *J* = 8.8 Hz, 1 H), [7.23 (d, *J* = 3.6 Hz), 7.18 (d, *J* = 3.6 Hz) (1 H)], 6.74 (br s, 1 H), [6.70 (d, *J* = 9.6 Hz), 6.66 (d, *J* = 9.2 Hz) (1 H)], 4.34 (q, *J* = 6.4 Hz, 2 H), 1.24 (t, *J* = 6.4 Hz, 3 H).

MS: *m*/*z* = 280.03 $[M + H]^+$.

Anal. Calcd for $C_{16}H_{13}N_3O_2$: C, 68.81; H, 4.69; N, 15.05. Found: C, 68.98; H, 4.65; N, 14.97.

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