An Efficient and Mild Synthesis of Tetrahydro-4*H*-indol-4-one Derivatives via a Domino Reaction in Water

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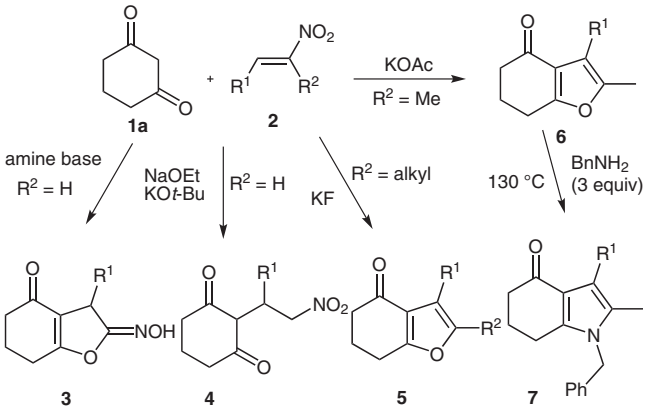
Abstract: An efficient domino approach for the synthesis of tetrahydro-4*H*-indol-4-one derivatives has been established. The use of catalytic amount of L-proline in water at 60 °C makes it quite simple, more convenient, and environmentally benign.

Key words: tetrahydro-4*H*-indol-4-one, amines, cyclization, domino reaction, catalysis

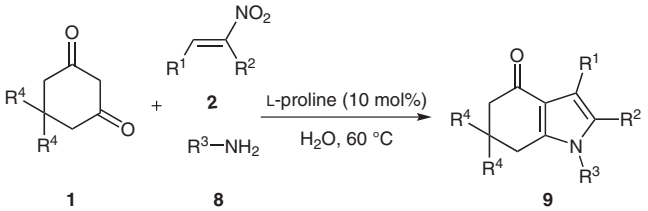
There is continuing interest in the development of improved methods for the synthesis of indole and its derivatives owing to their importance as 'privileged pharmacological structures' in natural bioactive products and marketed drugs. As a result, many powerful methodologies for the synthesis of these compounds have been developed,[1] the majority of them involve Fischer-type indole synthesis,[2] reductive cyclization,[1j,r] metal-catalyzed coupling/condensation cascades,[3] and electrophilic activation of *N*-aryl amides.[4] Recently, the utility of enamines in metal catalysis or under microwave irradiation for the formation of valuable indoles has also been reported.[5] In spite of a large number of methods reported for the synthesis of indole derivatives, there remains considerable demand to explore more convenient, practical, and benign reagents for their synthesis, especially for tetrahydroindoles that display a wide spectrum of biological activity and are important intermediates in the synthesis of highly functionalized indoles.[3d] Therefore, the development of concise approaches to indole and its derivatives by simple, convenient, and practical procedures and from inexpensive starting materials is of great importance and a challenging endeavor.

Recently, organocatalysis has become a major, intensively active field in chemical synthesis due to its environmental friendliness.[6] Particularly, L-proline, as a readily available commercial catalyst, has received much attention due to its dual role as a ligand and a catalyst.[7] Accordingly, the wide application range of L-proline and its derivatives has been described though various organic transformations with excellent yields, such as enamine-based direct asymmetric aldol,[8] Mannich,[9] Michael,[10] Diels–Alder,[11] α-amination,[12] and Knoevenagel-type reactions,[13] as well as unsymmetric Biginelli reactions.[14] In addition, organocatalytic domino cyclizations, where multiple C–C bond formations are achieved in a single operation using simple experimental procedures, have served as a powerful tool for the efficient assembly of complex structures with minimized waste production. Recently, L-proline and its derivatives have been used in domino cyclizations.[15] In view of the above perceptions, the development of a benign and organocatalytic domino cyclization process for the synthesis of tetrahydroindole derivatives is desirable.

Nitroolefins are building blocks for generating molecules of biological and pharmaceutical relevance.[16] Several groups have reported the base-dependent chemoselective reactions of cyclohexane-1,3-dione (**1a**) and nitroolefins **2**, and for the generation of bicyclic oximes **3**,[17] nitro compound **4**,[18c] or hydrobenzofuran **5** or **6**[18] (Scheme 1). In addition, Arai et al. also reported synthesis of 4-oxohydroindoles **7** from 4-oxohydrobenzofuran with excessive benzylamine at 130 °C.[18c]



Scheme 1 The reported reaction pathways between cyclohexane-1,3-dione and nitroalkenes



Scheme 2 Synthesis of tetrahydro-4*H*-indol-4-ones

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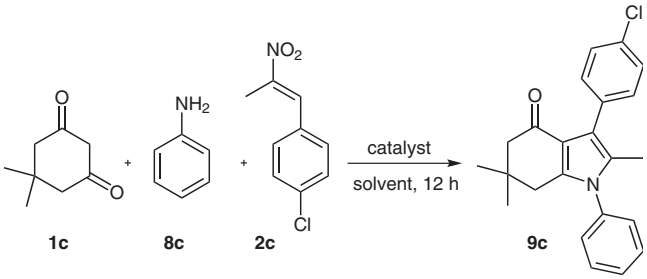
In continuation of our efforts to develop new methods for the synthesis of useful heterocyclic blocks from common starting materials,[19] herein, we wish to report a domino multicyclization reaction for the synthesis of new tetrahydro-4*H*-indol-4-one derivatives using L-proline as the catalyst in water (Scheme 2). Desired aromatic or aliphatic substituents can be introduced to 1- and 3-positions of the indole skeleton through this domino reaction. To the best of our knowledge, this convenient procedure for the efficient synthesis of tetrahydro-4*H*-indol-4-one derivatives using L-proline as the catalyst in water has not been previously reported in the literature.

We initially evaluated the domino reaction of 5,5-dimethylcyclohexane-1,3-dione (**1c**), nitroolefin **2c**, which was derived from the reaction of nitroethane with 4-chlorobenzaldehyde, and aniline **8c**. The reaction mixture, which was composed of a 1:1:1 mixture of **1c**, **2c**, and **8c**, was tested under various conditions; the results are presented in Table 1.

The optimization process revealed that none of the desired product was detected under catalyst-free conditions (Table 1, entry 1) and only a trace amount of product was observed, even at an enhanced temperature of 130 °C in toluene for 36 hours (entry 2). Interestingly, when the reaction was conducted in the presence of L-proline (10 mol%) as an abundant and inexpensive catalyst in ethanol, the desired product was obtained in 49% yield (entry 3). To improve the yield, the effect of the solvent was first studied and the results indicate that under the catalysis of L-proline (10 mol%) and heating at 60 °C for 12 hours in water, the desired product **9c** was isolated in 87% yield (entry 10). Similar results were observed when the solvent was changed to methanol and isopropyl alcohol (entries 8 and 9). However, poor yields were obtained when the reaction was carried out in non-protonic solvents, such as dichloromethane, *N*,*N*-dimethylformamide, tetrahydrofuran, and toluene. Then, the efficacies of several other organic and inorganic catalysts were examined for the reaction (entries 11–18); in all cases, 10 mol% of the catalyst was used and the reaction was carried out in water at 60 °C. L-Proline provided a superior catalytic effect to DL-alanine, 4-(dimethylamino)pyridine, triethylamine, pyrrolidine, *N*-methylmorpholine, pyridine, acetic acid, and potassium carbonate (entries 10–18), indicating that the presence of both secondary nitrogen and carboxylic acid group may be essential for better catalytic activity. Next, the effect of catalyst loading on the reaction was evaluated and the results revealed that increasing the amount of L-proline from 1 to 10 mol% led to an increase in the yield from 33% to 87% (entries 20, 21, and 10), but a further increase to 20% did not improve the yield significantly (entry 19). Finally, a strong dependence of the domino reaction on the temperature was observed. Upon varying the temperature of the reaction from 50 °C to room temperature, the yield also decline significantly (entries 24–26). When the temperature ranges from 40 to 50 °C, the reaction gave only poor yields of product **9c**, even after longer reaction times. Instead, the intermediate **10**, of

Table 1 Optimization of Reaction Conditions for the Synthesis of **9c**[a]



Entry	Solvent	Catalyst (mol%)	T (°C)	Yield[b] (%)
1	EtOH	–	60	–
2[c]	toluene	–	130	trace
3	EtOH	L-proline (10)	60	49
4	CH_2Cl_2	L-proline (10)	reflux	11
5	THF	L-proline (10)	60	31
6	DMF	L-proline (10)	60	25
7	toluene	L-proline (10)	60	25
8	*i*-PrOH	L-proline (10)	60	85
9	MeOH	L-proline (10)	60	88
10	H_2O	L-proline (10)	60	87
11	H_2O	DL-alanine (10)	60	69
12	H_2O	DMAP (10)	60	40
13	H_2O	Et_3N (10)	60	46
14	H_2O	pyrrolidine (10)	60	10
15	H_2O	NMM (10)	60	21
16	H_2O	pyridine (10)	60	29
17	H_2O	K_2CO_3 (10)	60	53
18	H_2O	AcOH	60	25
19	H_2O	L-proline (20)	60	88
20	H_2O	L-proline (5)	60	62
21	H_2O	L-proline (1)	60	33
22	H_2O	L-proline (10)	80	86
23	H_2O	L-proline (10)	reflux	85
24[c]	H_2O	L-proline (10)	50	29
25[c]	H_2O	L-proline (10)	40	18
26	H_2O	L-proline (10)	r.t.	–

[a] Reaction conditions: **1c** (0.5 mmol), **2c** (0.5 mmol), **8c** (0.5 mmol), solvent (3 mL), 12 h.
[b] Isolated yields.
[c] Reaction time: 36 h.

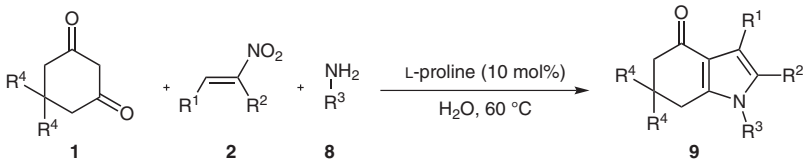
structure **D** (entries 24, 25 and Scheme 3), was observed in quantitative yield. When the reaction was performed in water at room temperature, no desired product was found. Instead, only hydrobenzofuran of the structure **6** was obtained. Additionally, it is noteworthy that when the R^2 substituent was H group, only bicyclic oxime **3** was obtained at room temperature. Moreover, the yield leveled off when the temperature was further increased to 80 and 100 °C (entries 22 and 23). Extensive screening showed that the optimal conditions involved the following parameters: 10 mol% L-proline as catalyst, and water as solvent, with the reaction temperature at 60 °C.

With optimal conditions established, we then examined the scope of the reaction for the construction of various tetrahydro-4*H*-indol-4-ones (Table 2). Initially, to test the scope of the nitroolefins, 5,5-dimethylcyclohexane-1,3-dione (**1c**) and aniline **8c** were used as model substrates, and the results indicate that a wide range of substituted groups of nitroolefins all gave the desired products in good to excellent yields, which include chloro, bromo, fluoro, nitro, methoxy, or methyl groups. In addition, no noticeable electronic effects were observed. It should be noted that good results were also obtained by using other aromatic systems, such as 1-nitro-2-thienylethene and 1-furyl-2-nitroethene (entries 8 and 9).

To further expand the scope of the amine substrates, we employed different nitroolefins and 5,5-dimethylcyclohexane-1,3-dione (**1c**) as model substrates and examined various amines, including aniline, 4-chloroaniline, 2-chloroaniline, *o*-toluidine, 2,4-dimethylaniline, 1-naphthylamine, cyclopropylamine cyclohexanamine, benzylamine, and pentan-1-amine. In all these cases, the reactions proceeded smoothly to give the appropriate expected products in good to excellent yields.

Finally, with a broad scope of nitroolefins and amines examined, our attention turned to using other ketones, such as cyclohexane-1,3-dione (**1a**) and pentane-2,4-dione (**1b**). When cyclohexane-1,3-dione (**1a**) was used under the optimized conditions, the reactions could be carried out smoothly to give the desired products with high yields (entries 25–39). Surprisingly, open-chain pentane-2,4-dione (**1b**) does not lead to the desired products by this procedure even after longer reaction times and higher temperatures.

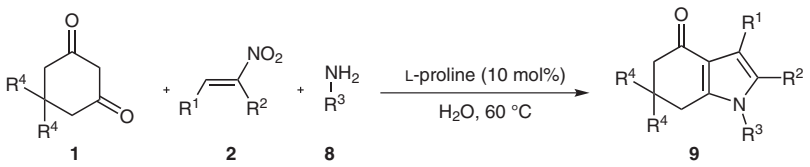
Table 2 Synthesis of **9** in Water[a]



1 2 8 9

Entry	9	R^1	R^2	R^3	R^4	Time (h)	Yield[b] (%)	Mp (°C)
1	**9a**	4-$O_2NC_6H_4$	Me	Ph	Me	12	67	106–108
2	**9b**	4-FC_6H_4	Me	Ph	Me	10	83	119–120
3	**9c**	4-ClC_6H_4	Me	Ph	Me	12	87	113–115
4	**9d**	4-BrC_6H_4	Me	Ph	Me	10	75	95–97
5	**9e**	Ph	Me	Ph	Me	8	83	140–142
6	**9f**	4-$MeOC_6H_4$	Me	Ph	Me	8	89	92–94
7	**9g**	4-MeC_6H_4	Me	Ph	Me	10	86	118–120
8	**9h**	2-thienyl	Me	Ph	Me	10	88	107–109
9	**9i**	2-furyl	Me	Ph	Me	10	80	149–150
10	**9j**	4-FC_6H_4	Me	4-ClC_6H_4	Me	12	76	184–186
11	**9k**	4-ClC_6H_4	Me	4-ClC_6H_4	Me	12	81	169–171
12	**9l**	Ph	Me	4-ClC_6H_4	Me	12	83	118–120
13	**9m**	4-$MeOC_6H_4$	Me	4-ClC_6H_4	Me	12	86	125–127
14	**9n**	4-MeC_6H_4	Me	4-ClC_6H_4	Me	12	79	136–138
15	**9o**	4-FC_6H_4	Me	2-MeC_6H_4	Me	12	75	113–115
16	**9p**	Ph	Me	2-MeC_6H_4	Me	12	83	96–98
17	**9q**	4-$MeOC_6H_4$	Me	2-MeC_6H_4	Me	12	86	109–111

Table 2 Synthesis of **9** in Water[a] (continued)

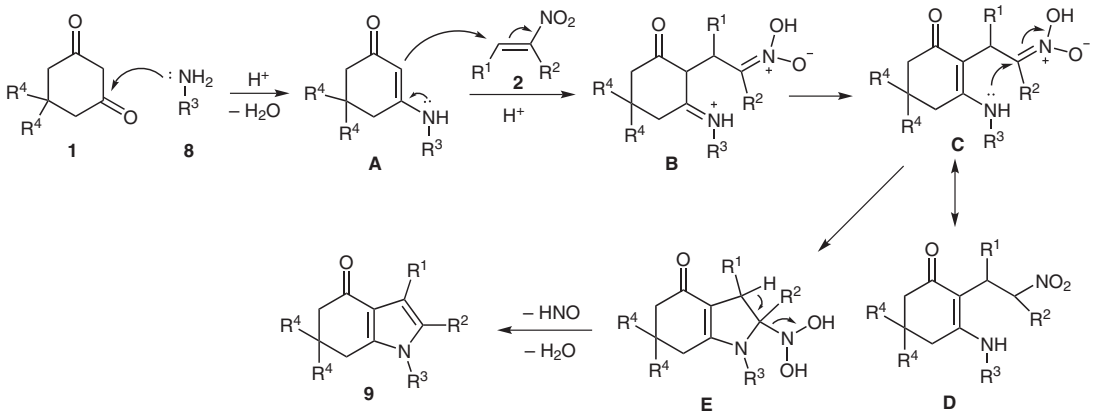


1 **2** **8** **9**

Entry	9	R^1	R^2	R^3	R^4	Time (h)	Yield[b] (%)	Mp (°C)
18	**9r**	4-MeC_6H_4	Me	2-MeC_6H_4	Me	12	82	121–123
19	**9s**	Ph	Me	2,4-$Me_2C_6H_3$	Me	12	86	110–112
20	**9t**	Ph	Me	2-$MeOC_6H_4$	Me	12	86	106–108
21	**9u**	4-ClC_6H_4	Me	1-naphthyl	Me	12	75	165–167
22	**9v**	Ph	Me	cyclopropyl	Me	12	76	148–149
23	**9w**	4-$MeOC_6H_4$	Me	cyclopropyl	Me	12	83	141–142
24	**9x**	Ph	H	Ph	Me	12	85	oil
25	**9y**	4-BrC_6H_4	Me	Ph	H	10	90	124–126
26	**9z**	Ph	Me	Ph	H	10	83	110–111
27	**9aa**	4-MeC_6H_4	Me	Ph	H	10	92	123–124
28	**9bb**	2-thienyl	Me	Ph	H	10	92	138–139
29	**9cc**	2-furyl	Me	Ph	H	10	70	143–145
30	**9dd**	Ph	Me	2-ClC_6H_4	H	10	79	99–101
31	**9ee**	4-$MeOC_6H_4$	Me	2-ClC_6H_4	H	10	85	106–108
32	**9ff**	Ph	Me	4-ClC_6H_4	H	10	81	164–165
33	**9gg**	4-ClC_6H_4	Me	2-MeC_6H_4	H	10	78	140–141
34	**9hh**	Ph	Me	2-MeC_6H_4	H	10	84	98–100
35	**9ii**	4-$MeOC_6H_4$	Me	2-MeC_6H_4	H	10	91	122–124
36	**9jj**	4-$O_2NC_6H_4$	Me	2,4-$Me_2C_6H_3$	H	10	84	134–136
37	**9kk**	Ph	Me	2,4-$Me_2C_6H_3$	H	10	87	128–130
38	**9ll**	Ph	Me	2-$MeOC_6H_4$	H	10	92	107–109
39	**9mm**	Ph	Me	1-naphthyl	H	10	77	127–129
40	**9nn**	Ph	Me	Cy	Me	12	83	oil
41	**9oo**	Ph	Me	Bn	Me	12	87	156–158
42	**9pp**	Ph	Me	$(CH_2)_4Me$	Me	12	85	oil

[a] Reaction conditions: **1** (0.5 mmol), **2** (0.5 mmol), **8** (0.5 mmol), L-proline (10 mol%), H_2O (3 mL), 60 °C.
[b] Isolated yields.

According to reported literatures,[20] a plausible mechanism for the domino cyclization was proposed (Scheme 3). An initial condensation generated enaminones **A**, which further undergoes in situ a conjugate addition of the nitroolefin **2** to give intermediate **B**. Intermediate **B** undergoes imine–enamine tautomerization to give radical intermediate **C** or **D**. Finally, intramolecular cyclization followed by elimination of H_2O and HNO from the intermediate **E** afford the desired product **9**. To test the proposed mechanism, intermediate of type **A** (R^3 = Ph, R^4 = Me) was separately prepared from 5,5-dimethylcyclohexane-1,3-dione and aniline and when a two-component coupling reaction of enaminones (intermediate **A**) and nitroolefin **1c** was performed under the same reaction conditions, the target product **9c** was obtained with a yield similar to that of the one-pot reaction. Additionally, intermediate **10**, of type **D**, was separated and indentified by various spectral data.



Scheme 3 Proposed mechanism

In conclusion, we have developed an L-proline-catalyzed procedure for the facile synthesis of a variety of potentially biologically active tetrahydro-4*H*-indol-4-ones based on a novel domino reaction. The ready accessibility of starting materials and the broad compatibility of nitroolefins and amines make these reactions highly valuable for organic and biomedical fields. The continuing work on this project will be focused on the development of asymmetric versions of these reactions.

All reagents were commercial products without further purification, unless otherwise stated. Analytical TLC was performed using Merck silica gel GF254 plates. Flash column chromatography was performed on silica gel (200–300 mesh). Melting points were measured on an X-4 melting point apparatus. IR spectra were recorded with a Nicolet Nexus 670 FT-IR spectrophotometer. ^{1}H NMR spectra were recorded on a 400 MHz instrument (Bruker Avance 400 Spectrometer) relative to TMS as the internal reference. ^{13}C NMR spectra were recorded at 100 MHz with the internal chloroform signal at δ = 77.0 as a standard. Elemental analysis was carried out on EuroEA elemental analyzer. ESI-MS was determined by using the LCQ Fleet HPLC/MS instrument (Thermo Finnigan). HRMS (ESI) was measured with a Bruker Daltonics APEXII instrument.

Tetrahydro-4*H*-indol-4-ones 9; General Procedure

In a 10-mL reaction vial, nitroolefin **2** (0.5 mmol), 1,3-dione **1** (0.5 mmol), amine **8** (0.5 mmol), L-proline (10 mol%), and H_2O (3.0 mL) were mixed and then capped. The mixture was stirred for the given time (Table 2) at 60 °C. Upon completion (TLC monitoring), the mixture was cooled to r.t. and extracted with EtOAc (3 × 5 mL). The resulting residue was purified by column chromatography (silica gel, EtOAc–petroleum ether, 1:20–1:5) to give the pure product.

2,6,6-Trimethyl-3-(4-nitrophenyl)-1-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9a)

Yellow solid; yield: 125.6 mg (67%); mp 106–108 °C.

IR (KBr): 2955, 1644, 1590, 1499, 1482, 1451, 1439, 1419, 1066, 1042, 955, 831, 752 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.27–8.20 (m, 4 H, H_{Ar}), 7.76 (d, *J* = 8.8 Hz, 1 H, H_{Ar}), 7.62–7.56 (m, 4 H, H_{Ar}), 2.46 (s, 2 H, CH_2), 2.41 (s, 2 H, CH_2), 2.04 (s, 3 H, CH_3), 1.10 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.3, 148.2, 143.9, 142.0, 136.7, 131.0, 129.8, 129.1, 127.7, 127.6, 124.0, 122.9, 116.2, 53.0, 37.0, 35.3, 28.5, 11.3.

MS (ESI): *m*/*z* (%) = 375.24 (100) $[M + H]^+$.

Anal. Calcd for $C_{23}H_{22}N_2O_3$: C, 73.78; H, 5.92; N, 7.48. Found: C, 73.46; H, 5.81; N, 7.61.

3-(4-Fluorophenyl)-2,6,6-trimethyl-1-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9b)

Yellow solid; yield: 144.3 mg (83%); mp 119–120 °C.

IR (KBr): 2945, 1645, 1591, 1491, 1485, 1067, 976, 833, 691 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.58–7.51 (m, 4 H, H_{Ar}), 7.43–7.39 (m, 2 H, H_{Ar}), 7.30 (s, 1 H, H_{Ar}), 7.07 (t, *J* = 8.8 Hz, 2 H, H_{Ar}), 2.45 (s, 2 H, CH_2), 2.38 (s, 2 H, CH_2), 1.99 (s, 3 H, CH_3), 1.09 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.1, 160.4, 142.9, 137.2, 131.9, 131.8, 130.5, 129.6, 128.8, 128.5, 127.8, 119.4, 116.4, 114.5, 114.3, 53.1, 37.0, 35.2, 28.5, 11.0.

MS (ESI): *m*/*z* (%) = 348.25 (100) $[M + H]^+$.

Anal. Calcd for $C_{23}H_{22}FNO$: C, 79.51; H, 6.38; N, 4.03. Found: C, 79.28; H, 6.49; N, 3.89.

3-(4-Chlorophenyl)-2,6,6-trimethyl-1-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9c)

Yellow solid; yield: 158.2 mg (87%); mp 113–115 °C.

IR (KBr): 2958, 1657, 1591, 1497, 1475, 1061, 995, 802, 721 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.58–7.50 (m, 4 H, H_{Ar}), 7.40–7.33 (m, 5 H, H_{Ar}), 2.44 (s, 2 H, CH_2), 2.38 (s, 2 H, CH_2), 2.00 (s, 3 H, CH_3), 1.09 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.2, 143.2, 137.1, 133.1, 132.0, 131.7, 129.6, 128.8, 128.7, 127.8, 127.7, 119.2, 116.3, 53.1, 37.0, 35.2, 28.5, 11.1.

HRMS (ESI): *m*/*z* $[M + H]^+$ calcd for $C_{23}H_{23}ClNO$: 364.1463; found: 364.1458.

3-(4-Bromophenyl)-2,6,6-trimethyl-1-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9d)

Yellow solid; yield: 153.1 mg (75%); mp 95–97 °C.

IR (KBr): 2955, 1651, 1597, 1456, 1431, 1057, 996, 852, 728 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.58–7.47 (m, 7 H, H_{Ar}), 7.32 (d, *J* = 8.0 Hz, 2 H, H_{Ar}), 2.44 (s, 2 H, CH_2), 2.38 (s, 2 H, CH_2), 1.99 (s, 3 H, CH_3), 1.09 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.2, 143.2, 137.1, 133.6, 132.1, 130.7, 129.7, 128.9, 128.7, 127.8, 120.2, 119.2, 116.2, 53.1, 37.0, 35.2, 28.5, 11.1.

MS (ESI): *m*/*z* (%) = 408.19 (100) $[M + H]^+$, 410.15.

Anal. Calcd for $C_{23}H_{22}BrNO$: C, 67.65; H, 5.43; N, 3.43. Found: C, 67.88; H, 5.39; N, 3.61.

2,6,6-Trimethyl-1,3-diphenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9e)
Yellow solid; yield: 137.3 mg (83%); mp 140–142 °C.

IR (KBr): 2968, 1651, 1485, 1481, 1112, 975, 889, 830, 738, 691 cm^{-1}.

^1H NMR (400 MHz, $CDCl_3$): δ = 7.58–7.51 (m, 3 H, H_{Ar}), 7.47–7.45 (m, 2 H, H_{Ar}), 7.41–7.36 (m, 2 H, H_{Ar}), 7.31–7.29 (m, 3 H, H_{Ar}), 2.46 (s, 2 H, CH_2), 2.39 (s, 2 H, CH_2), 2.02 (s, 3 H, CH_3), 1.10 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.1, 143.0, 137.3, 134.6, 130.4, 129.6, 128.7, 128.6, 127.8, 127.6, 126.2, 120.3, 116.4, 53.2, 37.1, 35.2, 28.5, 11.2.

MS (ESI): *m*/*z* (%) = 330.27 (100) $[M + H]^+$, 302.29, 216.28.

Anal. Calcd for $C_{23}H_{23}NO$: C, 83.85; H, 7.04; N, 4.25. Found: C, 83.53; H, 6.91; N, 4.58.

3-(4-Methoxyphenyl)-2,6,6-trimethyl-1-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9f)
Yellow solid; yield: 160.2 mg (89%); mp 92–94 °C.

IR (KBr): 2943, 1657, 1497, 1431, 1222, 1113, 975, 856, 731 cm^{-1}.

^1H NMR (400 MHz, $CDCl_3$): δ = 7.47–7.42 (m, 3 H, H_{Ar}), 7.30 (d, *J* = 8.4 Hz, 2 H, H_{Ar}), 7.22–7.21 (m, 2 H, H_{Ar}), 6.86 (d, *J* = 8.8 Hz, 2 H, H_{Ar}), 3.77 (s, 3 H, OCH_3), 2.36 (s, 2 H, CH_2), 2.30 (s, 2 H, CH_2), 1.93 (s, 3 H, CH_3), 1.00 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.2, 158.1, 142.8, 137.3, 131.4, 129.5, 128.6, 128.2, 127.8, 126.9, 119.9, 116.4, 113.1, 55.2, 53.2, 37.1, 35.2, 28.5, 11.1.

MS (ESI): *m*/*z* (%) = 360.24 (100) $[M + H]^+$, 274.36.

Anal. Calcd for $C_{24}H_{25}NO_2$: C, 80.19; H, 7.01; N, 3.90. Found: C, 80.52; H, 6.89; N, 4.07.

2,6,6-Trimethyl-1-phenyl-3-(*p*-tolyl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9g)
Yellow solid; yield: 147.7 mg (86%); mp 118–120 °C.

IR (KBr): 2958, 1655, 1459, 1432, 1130, 997, 853, 802, 729 cm^{-1}.

^1H NMR (400 MHz, $CDCl_3$): δ = 7.55–7.50 (m, 4 H, H_{Ar}), 7.36–7.30 (m, 3 H, H_{Ar}), 7.21–7.16 (m, 2 H, H_{Ar}), 2.45 (s, 2 H, CH_2), 2.39 (s, 3 H, CH_3), 2.37 (s, 2 H, CH_2), 2.01 (s, 3 H, CH_3), 1.09 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.9, 142.7, 137.4, 135.7, 131.6, 130.2, 129.5, 128.6, 128.3, 127.8, 120.3, 116.5, 111.4, 53.2, 37.1, 35.1, 28.5, 21.2, 11.1.

HRMS (ESI): *m*/*z* $[M + H]^+$ calcd for $C_{24}H_{26}NO$: 344.2009; found: 344.2003.

2,6,6-Trimethyl-1-phenyl-3-(thiophen-2-yl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9h)
Yellow solid; yield: 147.8 mg (88%); mp 107–109 °C.

IR (KBr): 2938, 1649, 1487, 1321, 1221, 1158, 821, 739, 695 cm^{-1}.

^1H NMR (400 MHz, $CDCl_3$): δ = 7.54–7.51 (m, 4 H, H_{Ar}), 7.33–7.28 (m, 4 H, H_{Ar}), 2.42 (s, 2 H, CH_2), 2.37 (s, 2 H, CH_2), 2.06 (s, 3 H, CH_3), 1.07 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.2, 142.9, 137.2, 134.5, 130.3, 129.6, 128.8, 128.7, 127.8, 124.8, 123.5, 122.7, 116.5, 53.2, 37.1, 35.1, 29.7, 28.5, 11.5.

MS (ESI): *m*/*z* (%) = 336.22 (100) $[M + H]^+$, 274.33, 266.31, 231.23.

Anal. Calcd for $C_{21}H_{21}NOS$: C, 75.19; H, 6.31; N, 4.18. Found: C, 75.40; H, 6.37; N, 4.39.

3-(Furan-2-yl)-2,6,6-trimethyl-1-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9i)
Brown solid; yield: 128.2 mg (80%); mp 149–150 °C.

IR (KBr): 2940, 1655, 1456, 1328, 1056, 975, 821, 734, 698 cm^{-1}.

^1H NMR (400 MHz, $CDCl_3$): δ = 7.54–7.49 (m, 3 H, H_{Ar}), 7.43 (s, 1 H, H_{Ar}), 7.23–7.21 (m, 2 H, H_{Ar}), 7.10 (d, *J* = 3.2 Hz, 1 H, H_{Ar}), 6.48–6.46 (m, 1 H, H_{Ar}), 2.39 (s, 2 H, CH_2), 2.36 (s, 2 H, CH_2), 2.21 (s, 3 H, CH_3), 1.05 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.8, 140.5, 132.6, 129.7, 129.6, 128.9, 128.0, 127.9, 121.5, 115.8, 111.0, 109.2, 53.3, 37.1, 34.9, 28.4, 12.1.

MS (ESI): *m*/*z* (%) = 320.24 (100) $[M + H]^+$.

Anal. Calcd for $C_{21}H_{21}NO_2$: C, 78.97; H, 6.63; N, 4.39. Found: C, 79.16; H, 6.49; N, 4.18.

1-(4-Chlorophenyl)-3-(4-fluorophenyl)-2,6,6-trimethyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9j)
Yellow solid; yield: 145.2 mg (76%); mp 184–186 °C.

IR (KBr): 2941, 1649, 1457, 1321, 1103, 975, 887, 729, 679 cm^{-1}.

^1H NMR (400 MHz, $CDCl_3$): δ = 7.55–7.53 (m, 2 H, H_{Ar}), 7.40–7.37 (m, 2 H, H_{Ar}), 7.25–7.23 (m, 2 H, H_{Ar}), 7.09–7.05 (m, 2 H, H_{Ar}), 2.44 (s, 2 H, CH_2), 2.38 (s, 2 H, CH_2), 1.99 (s, 3 H, CH_3), 1.09 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.2, 142.8, 135.6, 134.9, 131.9, 131.8, 130.2, 129.9, 129.1, 128.4, 119.7, 116.6, 114.6, 114.4, 53.1, 37.0, 35.2, 28.5, 11.0.

MS (ESI): *m*/*z* (%) = 382.22 (100) $[M + H]^+$.

Anal. Calcd for $C_{23}H_{21}ClFNO$: C, 72.34; H, 5.54; N, 3.67. Found: C, 72.09; H, 5.43; N, 3.58.

1,3-Bis(4-chlorophenyl)-2,6,6-trimethyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9k)
Yellow solid; yield: 161.2 mg (81%); mp 169–171 °C.

IR (KBr): 2940, 1643, 1487, 1223, 988, 851, 739, 669 cm^{-1}.

^1H NMR (400 MHz, $CDCl_3$): δ = 7.55–7.52 (m, 2 H, H_{Ar}), 7.38–7.35 (m, 4 H, H_{Ar}), 7.27–7.23 (m, 2 H, H_{Ar}), 2.43 (s, 2 H, CH_2), 2.37 (s, 2 H, CH_2), 1.99 (s, 3 H, CH_3), 1.09 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.1, 143.0, 135.6, 134.9, 132.8, 132.1, 131.7, 129.9, 129.2, 129.1, 128.6, 127.8, 125.4, 119.5, 116.5, 53.0, 37.0, 35.2, 28.5, 11.1.

MS (ESI): *m*/*z* (%) = 398.23 (100) $[M + H]^+$, 362.41, 324.36.

Anal. Calcd for $C_{23}H_{21}Cl_2NO$: C, 69.35; H, 5.31; N, 3.52. Found: C, 69.26; H, 5.01; N, 3.61.

1-(4-Chlorophenyl)-2,6,6-trimethyl-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9l)
Yellow solid; yield: 150.8 mg (83%); mp 118–120 °C.

IR (KBr): 2956, 1658, 1458, 1433, 1228, 975, 837, 809, 726, 684 cm^{-1}.

^1H NMR (400 MHz, $CDCl_3$): δ = 7.55–7.52 (m, 2 H, H_{Ar}), 7.44–7.36 (m, 5 H, H_{Ar}), 7.29–7.24 (m, 2 H, H_{Ar}), 2.44 (s, 2 H, CH_2), 2.38 (s, 2 H, CH_2), 2.01 (s, 3 H, CH_3), 1.09 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.0, 142.7, 135.8, 134.8, 134.3, 130.4, 129.9, 129.1, 128.7, 128.4, 127.6, 126.3, 120.7, 116.7, 53.1, 37.1, 35.2, 28.5, 11.1.

MS (ESI): *m*/*z* (%) = 364.21 (100) $[M + H]^+$, 225.18.

Anal. Calcd for $C_{23}H_{22}ClNO$: C, 75.92; H, 6.09; N, 3.85. Found: C, 75.56; H, 6.00; N, 3.92.

1-(4-Chlorophenyl)-3-(4-methoxyphenyl)-2,6,6-trimethyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9m)
Yellow solid; yield: 169.2 mg (86%); mp 125–127 °C.

IR (KBr): 2960, 1651, 1436, 1358, 1201, 995, 872, 731, 687 cm^{-1}.

^1H NMR (400 MHz, $CDCl_3$): δ = 7.53 (d, *J* = 8.4 Hz, 2 H, H_{Ar}), 7.36 (d, *J* = 8.8 Hz, 2 H, H_{Ar}), 7.24 (d, *J* = 8.4 Hz, 2 H, H_{Ar}), 6.94 (d,

J = 8.8 Hz, 2 H, H_{Ar}), 3.85 (s, 3 H, OCH_3), 2.43 (s, 2 H, CH_2), 2.37 (s, 2 H, CH_2), 2.00 (s, 3 H, CH_3), 1.09 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.1, 158.1, 149.7, 145.2, 142.6, 135.8, 131.4, 129.8, 129.1, 116.7, 113.1, 55.2, 53.1, 37.1, 35.2, 28.5, 11.1.

MS (ESI): *m*/*z* (%) = 394.24 (100) $[M + H]^+$, 360.32, 274.33.

Anal. Calcd for $C_{24}H_{24}ClNO_2$: C, 73.18; H, 6.14; N, 3.56. Found: C, 73.40; H, 6.32; N, 3.74.

1-(4-Chlorophenyl)-2,6,6-trimethyl-3-(*p*-tolyl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9n)
Yellow solid; yield: 149.2 mg (79%); mp 136–138 °C.

IR (KBr): 2958, 1659, 1499, 1431, 1325, 1226, 1201, 997, 821, 777, 726 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.54–7.52 (m, 2 H, H_{Ar}), 7.32 (d, *J* = 8.0 Hz, 2 H, H_{Ar}), 7.24 (d, *J* = 8.8 Hz, 2 H, H_{Ar}), 7.19 (d, *J* = 7.6 Hz, 2 H, H_{Ar}), 2.43 (s, 2 H, CH_2), 2.38 (s, 3 H, OCH_3), 2.37 (s, 2 H, CH_2), 2.00 (s, 3 H, CH_3), 1.09 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.0, 142.6, 135.9, 135.8, 134.7, 131.3, 130.2, 129.8, 129.1, 128.4, 128.2, 120.6, 116.7, 53.1, 37.1, 35.2, 28.5, 21.3, 11.1.

HRMS (ESI): *m*/*z* $[M + H]^+$ calcd for $C_{24}H_{25}ClNO$: 378.1619; found: 378.1615.

3-(4-Fluorophenyl)-2,6,6-trimethyl-1-(*o*-tolyl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9o)
Yellow solid; yield: 135.6 mg (75%); mp 113–115 °C.

IR (KBr): 2956, 1658, 1597, 1523, 1497, 1460, 1385, 1222, 1158, 1111, 825, 775, 729 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.44–7.35 (m, 5 H, H_{Ar}), 7.21 (d, *J* = 7.6 Hz, 1 H, H_{Ar}), 7.07 (t, *J* = 8.8 Hz, 2 H, H_{Ar}), 2.44–2.32 (m, 3 H, CH_2), 2.19–2.15 (m, 1 H, CH_2), 2.05 (s, 3 H, CH_3), 1.90 (s, 3 H, CH_3), 1.09 (d, *J* = 4.0 Hz, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.1, 142.8, 136.3, 136.2, 131.9, 131.2, 129.4, 128.4, 128.2, 127.2, 116.2, 114.5, 114.3, 111.2, 53.2, 36.7, 35.2, 29.0, 28.0, 17.2, 10.5.

MS (ESI): *m*/*z* (%) = 362.23 (100) $[M + H]^+$, 243.19.

Anal. Calcd for $C_{24}H_{24}FNO$: C, 79.75; H, 6.69; N, 3.88. Found: C, 79.46; H, 6.77; N, 3.69.

2,6,6-Trimethyl-3-phenyl-1-(*o*-tolyl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9p)
Yellow solid; yield: 142.8 mg (83%); mp 96–98 °C.

IR (KBr): 2955, 1657, 1469, 1452, 1324, 1221, 977, 833, 801, 696 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.48–7.37 (m, 7 H, H_{Ar}), 7.29–7.21 (d, *J* = 7.6 Hz, 2 H, H_{Ar}), 2.45–2.32 (m, 3 H, CH_2), 2.19–2.16 (m, 1 H, CH_2), 2.05 (s, 3 H, CH_3), 1.92 (s, 3 H, CH_3), 1.09 (d, *J* = 4.0 Hz, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.0, 150.4, 142.8, 136.4, 136.3, 134.6, 131.2, 130.4, 129.3, 128.4, 127.5, 127.2, 126.1, 120.2, 116.3, 53.2, 36.7, 35.2, 29.1, 28.0, 17.3, 10.6.

MS (ESI): *m*/*z* (%) = 344.28 (100) $[M + H]^+$.

Anal. Calcd for $C_{24}H_{25}NO$: C, 83.93; H, 7.34; N, 4.08. Found: C, 83.78; H, 7.21; N, 3.98.

3-(4-Methoxyphenyl)-2,6,6-trimethyl-1-(*o*-tolyl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9q)
Yellow solid; yield: 160.6 mg (86%); mp 109–111 °C.

IR (KBr): 2955, 1648, 1589, 1531, 1490, 1457, 1375, 1231, 1150, 1118, 830, 766, 728 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.43–7.35 (m, 5 H, H_{Ar}), 7.21 (d, *J* = 7.6 Hz, 1 H, H_{Ar}), 6.94 (d, *J* = 8.8 Hz, 2 H, H_{Ar}), 3.85 (s, 3 H, OCH_3), 2.44–2.32 (m, 3 H, CH_2), 2.26–2.19 (m, 1 H, CH_2), 2.05 (s, 3 H, CH_3), 1.91 (s, 3 H, CH_3), 1.08 (d, *J* = 3.6 Hz, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.1, 158.0, 142.6, 136.4, 136.3, 131.4, 131.1, 129.3, 128.4, 127.8, 127.2, 127.0, 119.7, 116.2, 113.1, 55.2, 53.2, 36.7, 35.1, 29.1, 27.9, 17.3, 10.6.

MS (ESI): *m*/*z* (%) = 374.31 (100) $[M + H]^+$, 318.34, 230.27.

Anal. Calcd for $C_{25}H_{27}NO_2$: C, 80.40; H, 7.29; N, 3.75. Found: C, 80.03; H, 7.36; N, 3.84.

2,6,6-Trimethyl-1-(*o*-tolyl)-3-(*p*-tolyl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9r)
Yellow solid; yield: 146.5 mg (82%); mp 121–123 °C.

IR (KBr): 2953, 1657, 1552, 1496, 1459, 1413, 1385, 1353, 1299, 1113, 1058, 828, 772, 729 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.42–7.34 (m, 4 H, H_{Ar}), 7.21–7.18 (m, 4 H, H_{Ar}), 2.43–2.29 (m, 6 H, CH_2, CH_3), 2.18–2.13 (m, 1 H, CH_2), 2.00 (s, 3 H, CH_3), 1.90 (s, 3 H, CH_3), 1.08 (d, *J* = 4.0 Hz, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.9, 142.6, 136.3, 135.6, 131.1, 130.2, 129.5, 129.3, 128.4, 128.3, 128.0, 127.1, 120.1, 116.3, 53.2, 36.7, 35.1, 29.1, 27.9, 21.3, 17.3, 10.6.

MS (ESI): *m*/*z* (%) = 358.25 (100) $[M + H]^+$, 274.31, 239.22.

Anal. Calcd for $C_{25}H_{27}NO$: C, 83.99; H, 7.61; N, 3.92. Found: C, 84.17; H, 7.40; N, 4.13.

1-(2,4-Dimethylphenyl)-2,6,6-trimethyl-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9s)
Yellow solid; yield: 153.8 mg (86%); mp 110–112 °C.

IR (KBr): 2958, 1652, 1549, 1493, 1451, 1428, 1387, 1346, 1295, 1118, 1060, 820, 728 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.46 (d, *J* = 6.8 Hz, 2 H, H_{Ar}), 7.39–7.35 (m, 3 H, H_{Ar}), 7.19 (s, 1 H, H_{Ar}), 7.15 (d, *J* = 8.4 Hz, 1 H, H_{Ar}), 7.08 (d, *J* = 8.0 Hz, 1 H, H_{Ar}), 2.43–2.34 (m, 7 H, CH_2, CH_3), 1.99 (s, 3 H, CH_3), 1.91 (s, 3 H, CH_3), 1.07 (d, *J* = 4.4 Hz, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.0, 143.0, 139.3, 135.9, 134.7, 133.7, 131.8, 131.4, 130.4, 128.7, 128.4, 128.1, 127.8, 127.5, 127.0, 116.1, 53.2, 36.7, 35.2, 29.1, 28.0, 21.2, 17.2, 10.6.

MS (ESI): *m*/*z* (%) = 358.30 (100) $[M + H]^+$.

Anal. Calcd for $C_{25}H_{27}NO$: C, 83.99; H, 7.61; N, 3.92. Found: C, 83.62; H, 7.56; N, 3.71.

1-(2-Methoxyphenyl)-2,6,6-trimethyl-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9t)
Yellow solid; yield: 154.8 mg (86%); mp 106–108 °C.

IR (KBr): 2949, 1649, 1541, 1497, 1457, 1420, 1380, 1344, 1295, 1063, 827, 720 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.50–7.45 (m, 3 H, H_{Ar}), 7.37 (t, *J* = 8.0 Hz, 2 H, H_{Ar}), 7.26–7.21 (m, 2 H, H_{Ar}), 7.11–7.07 (m, 2 H, H_{Ar}), 3,82 (s, 3 H, OCH_3), 2.41–2.29 (m, 4 H, CH_2), 1.94 (s, 3 H, CH_3), 1.08 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.1, 155.4, 143.9, 134.8, 130.5, 130.4, 129.7, 129.0, 127.4, 126.0, 125.8, 120.9, 119.8, 116.2, 112.2, 55.7, 36.6, 35.1, 28.8, 28.3, 10.6.

MS (ESI): *m*/*z* (%) = 360.27 (100) $[M + H]^+$, 274.39, 246.22.

Anal. Calcd for $C_{24}H_{25}NO_2$: C, 80.19; H, 7.01; N, 3.90. Found: C, 80.37; H, 7.33; N, 3.81.

3-(4-Chlorophenyl)-2,6,6-trimethyl-1-(naphthalen-1-yl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9u)
Red solid; yield: 155.2 mg (75%); mp 165–167 °C.

IR (KBr): 2956, 1658, 1596, 1526, 1491, 1430, 1422, 1386, 1367, 1351, 1090, 1013, 966, 826, 742 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.05–8.00 (m, 2 H, H_{Ar}), 7.65–7.54 (m, 4 H, H_{Ar}), 7.48–7.45 (m, 3 H, H_{Ar}), 7.37 (d, *J* = 8.4 Hz, 2 H, H_{Ar}), 2.45–2.35 (m, 3 H, CH_2), 2.14–2.10 (m, 1 H, CH_2), 1.88 (s, 3 H, CH_3), 1.05 (s, 3 H, CH_3), 1.02 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.2, 144.3, 134.3, 133.7, 133.1, 132.0, 131.8, 130.9, 129.7, 129.0, 128.5, 128.2, 127.9, 127.7, 127.1, 126.1, 125.4, 122.4, 119.1, 116.3, 53.2, 36.4, 35.1, 28.9, 27.8, 10.5.

MS (ESI): *m*/*z* (%) = 414.24 (100) $[M + H]^+$.

Anal. Calcd for $C_{27}H_{24}ClNO$: C, 78.34; H, 5.84; N, 3.38. Found: C, 78.11; H, 5.97; N, 3.29.

1-Cyclopropyl-2,6,6-trimethyl-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9v)
White solid; yield: 111.6 mg (76%); mp 148–149 °C.

IR (KBr): 2955, 1655, 1543, 1499, 1457, 1426, 1390, 1344, 1289, 1112, 1058, 821, 697 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.39–7.33 (m, 4 H, H_{Ar}), 7.27–7.24 (m, 1 H, H_{Ar}), 3.02–2.99 (m, 1 H, CH), 2.79 (s, 2 H, CH_2), 2.32 (s, 2 H, CH_2), 2.28 (s, 3 H, CH_3), 1.16–1.14 (m, 8 H, CH_2, CH_3), 0.99–0.98 (m, 2 H, CH_2).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.8, 144.1, 134.8, 130.4, 129.9, 127.5, 126.1, 120.0, 115.9, 52.9, 37.6, 35.1, 28.7, 26.0, 11.3, 7.6.

HRMS (ESI): *m*/*z* $[M + H]^+$ calcd for $C_{20}H_{24}NO$: 294.1852; found: 294.1848.

1-Cyclopropyl-3-(4-methoxyphenyl)-2,6,6-trimethyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9w)
Yellow solid; yield: 134.4 mg (83%); mp 141–142 °C.

IR (KBr): 2958, 1655, 1550, 1499, 1457, 1421, 1380, 1343, 1298, 1118, 829, 699 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.30–7.27 (m, 2 H, H_{Ar}), 6.92–6.90 (m, 2 H, H_{Ar}), 3.83 (s, 3 H, OCH_3), 3.02–2.97 (m, 1 H, CH), 2.78 (s, 2 H, CH_2), 2.31 (s, 2 H, CH_2), 2.26 (s, 3 H, CH_3), 1.15–1.13 (m, 8 H, CH_3, CH_2), 0.99–0.95 (m, 2 H, CH_2).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.9, 157.9, 143.9, 131.4, 129.5, 128.4, 127.1, 119.6, 115.9, 114.2, 113.0, 55.2, 52.9, 37.6, 35.1, 28.7, 26.0, 11.3, 7.6.

MS (ESI): *m*/*z* (%) = 324.28 (100) $[M + H]^+$, 274.40, 196.25.

Anal. Calcd for $C_{21}H_{25}NO_2$: C, 77.98; H, 7.79; N, 4.33. Found: C, 78.29; H, 7.83; N, 4.19.

6,6-Dimethyl-1,3-diphenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9x)
Yellow oil; yield: 134.2 mg (85%).

IR (KBr): 2944, 1641, 1547, 1492, 1438, 1429, 1388, 1351, 1299, 1119, 1057, 728, 698 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.71–7.69 (m, 2 H, H_{Ar}), 7.55–7.52 (m, 2 H, H_{Ar}), 7.47–7.43 (m, 2 H, H_{Ar}), 7.39–7.35 (m, 4 H, H_{Ar}), 6.91 (s, 1 H, CH), 2.68 (s, 2 H, CH_2), 2.46 (s, 2 H, CH_2), 1.12 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.6, 143.4, 138.5, 133.9, 129.6, 128.9, 128.7, 128.0, 127.9, 126.7, 126.6, 125.3, 121.9, 117.2, 53.2, 37.4, 35.3, 28.4.

MS (ESI): *m*/*z* (%) = 316.25 (100) $[M + H]^+$.

Anal. Calcd for $C_{22}H_{21}NO$: C, 83.78; H, 6.71; N, 4.44. Found: C, 83.99; H, 6.62; N, 4.31.

3-(4-Bromophenyl)-2-methyl-1-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9y)
Yellow solid; yield: 171.0 mg (90%); mp 124–126 °C.

IR (KBr): 2943, 1648, 1593, 1500, 1488, 1455, 1437, 1417, 1125, 1069, 1004, 930, 831, 759, 696 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.57–7.49 (m, 5 H, H_{Ar}), 7.32–7.27 (m, 4 H, H_{Ar}), 2.59 (t, *J* = 6.4 Hz, 2 H, CH_2), 2.50 (t, *J* = 6.4 Hz, 2 H, CH_2), 2.13–2.06 (m, 2 H, CH_2), 1.99 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.7, 144.3, 137.0, 133.7, 132.1, 130.7, 129.6, 128.9, 128.6, 127.7, 120.3, 119.3, 117.3, 39.0, 23.6, 23.0, 11.0.

MS (ESI): *m*/*z* (%) = 380.19 (100) $[M + H]^+$, 382.12.

Anal. Calcd for $C_{21}H_{18}BrNO$: C, 66.33; H, 4.77; N, 3.68. Found: C, 66.59; H, 4.69; N, 3.76.

2-Methyl-1,3-diphenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9z)
Yellow solid; yield: 125.2 mg (83%); mp 110–111 °C.

IR (KBr): 2960, 1728, 1657, 1445, 1367, 1335, 1288, 1205, 1115, 1072, 974, 910, 745 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.57–7.50 (m, 3 H, H_{Ar}), 7.48–7.43 (m, 4 H, H_{Ar}), 7.33–7.27 (m, 3 H, H_{Ar}), 2.60 (t, *J* = 6.0 Hz, 2 H, CH_2), 2.52–2.49 (m, 2 H, CH_2), 2.14–2.09 (m, 2 H, CH_2), 2.02 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.7, 144.0, 137.2, 134.7, 130.4, 129.6, 128.7, 128.4, 127.8, 127.6, 126.3, 120.4, 117.5, 39.1, 23.7, 23.1, 11.1.

MS (ESI): *m*/*z* (%) = 302.24 (100) $[M + H]^+$.

Anal. Calcd for $C_{21}H_{19}NO$: C, 83.69; H, 6.35; N, 4.65. Found: C, 83.51; H, 6.24; N, 4.71.

2-Methyl-1-phenyl-3-(*p*-tolyl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9aa)
Yellow solid; yield: 145.3 mg (92%); mp 123–124 °C.

IR (KBr): 2963, 1721, 1669, 1444, 1365, 1321, 1291, 1207, 1119, 1023, 974, 911, 725 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.56–7.49 (m, 3 H, H_{Ar}), 7.35–7.27 (m, 4 H, H_{Ar}), 7.20 (d, *J* = 8.0 Hz, 2 H, H_{Ar}), 2.59 (t, *J* = 6.0 Hz, 2 H, CH_2), 2.52–2.48 (m, 2 H, CH_2), 2.39 (s, 3 H, CH_3), 2.13–2.06 (m, 2 H, CH_2), 2.01 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.6, 143.9, 137.3, 135.8, 131.6, 130.2, 129.5, 128.7, 128.4, 128.2, 127.7, 120.3, 117.6, 39.1, 23.7, 23.1, 21.3, 11.1.

MS (ESI): *m*/*z* (%) = 316.29 (100) $[M + H]^+$, 301.30, 188.20.

Anal. Calcd for $C_{22}H_{21}NO$: C, 83.78; H, 6.71; N, 4.44. Found: C, 83.85; H, 6.83; N, 4.30.

2-Methyl-1-phenyl-3-(thiophen-2-yl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9bb)
Yellow solid; yield: 141.6 mg (92%); mp 138–139 °C.

IR (KBr): 2949, 1704, 1659, 1442, 1361, 1338, 1289, 1208, 1109, 1067, 937, 745 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.57–7.50 (m, 3 H, H_{Ar}), 7.32–7.27 (m, 5 H, H_{Ar}), 2.59–2.56 (m, 2 H, CH_2), 2.52–2.49 (m, 2 H, CH_2), 2.12–2.06 (m, 5 H, CH_2, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.8, 144.0, 137.2, 134.6, 130.4, 129.6, 128.8, 128.7, 127.7, 123.6, 122.7, 117.7, 115.1, 39.1, 23.6, 23.1, 11.5.

HRMS (ESI): *m*/*z* $[M + H]^+$ calcd for $C_{19}H_{18}NOS$: 308.1104; found: 308.1099.

3-(Furan-2-yl)-2-methyl-1-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9cc)
Brown solid; yield: 102.2 mg (70%); mp 143–145 °C.

IR (KBr): 2966, 1728, 1647, 1453, 1359, 1335, 1258, 1211, 1136, 1069, 972,729 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.57–7.50 (m, 3 H, H_{Ar}), 7.46–7.45 (m, 1 H, H_{Ar}), 7.25–7.27 (s, 2 H, H_{Ar}), 7.07–7.06 (m, 1 H, H_{Ar}),

6.50–6.48 (m, 1 H, H_{Ar}), 2.56–2.52 (m, 4 H, CH_2), 2.22 (s, 3 H, CH_3), 2.10–2.06 (m, 2 H, CH_2).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.3, 149.3, 146.2, 144.3, 140.6, 136.9, 129.6, 128.9, 127.9, 120.6, 111.0, 109.2, 39.2, 23.4, 23.2, 12.0.

MS (ESI): *m*/*z* (%) = 292.13 (100) $[M + H]^+$, 266.11.

Anal. Calcd for $C_{19}H_{17}NO_2$: C, 78.33; H, 5.88; N, 4.81. Found: C, 78.63; H, 5.97; N, 4.89.

1-(2-Chlorophenyl)-2-methyl-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9dd)
Yellow solid; yield: 132.8 mg (79%); mp 99–101 °C.

IR (KBr): 2958, 1657, 1456, 1366, 1331, 1279, 1215, 1119, 1067, 973, 910, 730 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.63–7.61 (m, 1 H, H_{Ar}), 7.49–7.44 (m, 4 H, H_{Ar}), 7.40–7.37 (m, 3 H, H_{Ar}), 7.30–7.29 (m, 1 H, H_{Ar}), 2.52–2.48 (m, 4 H, CH_2), 2.16–2.08 (m, 2 H, CH_2), 1.95 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.6, 144.1, 135.1, 134.6, 130.6, 130.4, 130.1, 128.3, 128.0, 127.5, 126.3, 120.4, 117.8, 39.1, 23.5, 22.5, 10.5.

MS (ESI): *m*/*z* (%) = 336.19 (100) $[M + H]^+$, 284.38, 222.15.

Anal. Calcd for $C_{21}H_{18}ClNO$: C, 75.11; H, 5.40; N, 4.17. Found: C, 74.87; H, 5.29; N, 4.42.

1-(2-Chlorophenyl)-3-(4-methoxyphenyl)-2-methyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9ee)
Yellow solid; yield: 155.6 mg (85%); mp 106–108 °C.

IR (KBr): 2961, 1658, 1440, 1355, 1337, 1289, 1201, 1105, 1070, 974, 726 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.53–7.50 (m, 1 H, H_{Ar}), 7.38–7.35 (m, 2 H, H_{Ar}), 7.30–7.28 (m, 3 H, H_{Ar}), 6.84 (d, *J* = 8.4 Hz, 2 H, H_{Ar}), 3.75 (s, 3 H, OCH_3), 2.41–2.38 (m, 4 H, CH_2), 2.03–2.00 (m, 2 H, CH_2), 1.84 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.7, 158.1, 144.0, 135.2, 133.4, 131.5, 130.6, 130.5, 130.1, 128.0, 127.9, 126.9, 117.8, 113.1, 55.2, 39.1, 23.5, 22.5, 10.5.

MS (ESI): *m*/*z* (%) = 366.26 (100) $[M + H]^+$, 274.38, 220.31.

Anal. Calcd for $C_{22}H_{20}ClNO_2$: C, 72.23; H, 5.51; N, 3.83. Found: C, 72.28; H, 5.63; N, 3.69.

1-(4-Chlorophenyl)-2-methyl-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9ff)
Yellow solid; yield: 136.0 mg (81%); mp 164–165 °C.

IR (KBr): 2951, 1655, 1450, 1366, 1339, 1289, 1201, 1082, 973, 917, 728 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.54–7.52 (m, 2 H, H_{Ar}), 7.43–7.36 (m, 4 H, H_{Ar}), 7.31–7.26 (m, 3 H, H_{Ar}), 2.59 (t, *J* = 6.0 Hz, 2 H, CH_2), 2.50 (t, *J* = 6.4 Hz, 2 H, CH_2), 2.14–2.08 (m, 2 H, CH_2), 2.01 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.6, 143.8, 135.7, 134.8, 134.4, 130.3, 129.8, 129.1, 128.3, 127.6, 126.4, 120.8, 117.8, 39.0, 23.6, 23.1, 11.0.

HRMS (ESI): *m*/*z* $[M + H]^+$ calcd for $C_{21}H_{19}ClNO$: 336.1150; found: 336.1145.

3-(4-Chlorophenyl)-2-methyl-1-(*o*-tolyl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9gg)
Yellow solid; yield: 136.4 mg (78%); mp 140–141 °C.

IR (KBr): 2956, 1721, 1652, 1446, 1359, 1289, 1203, 1119, 1070, 973, 900, 694 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.43–7.33 (m, 7 H, H_{Ar}), 7.22 (d, *J* = 7.6 Hz, 1 H, H_{Ar}), 2.56–2.48 (m, 3 H, CH_2), 2.40–2.31 (m, 1 H, CH_2), 2.13–2.07 (m, 2 H, CH_2), 2.05 (s, 3 H, CH_3), 1.90 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.7, 144.0, 136.3, 136.2, 133.2, 131.7, 131.2, 129.4, 128.3, 127.7, 127.2, 119.1, 117.3, 39.1, 23.6, 22.6, 17.3, 10.5.

HRMS (ESI): *m*/*z* $[M + H]^+$ calcd for $C_{22}H_{21}ClNO$: 350.1306; found: 350.1301.

2-Methyl-3-phenyl-1-(*o*-tolyl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9hh)
Yellow solid; yield: 132.6 mg (84%); mp 98–100 °C.

IR (KBr): 2952, 1721, 1652, 1446, 1358, 1340, 1279, 1206, 1118, 1070, 970, 685 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.47–7.38 (m, 7 H, H_{Ar}), 7.29–7.23 (m, 2 H, H_{Ar}), 2.51–2.48 (m, 2 H, CH_2), 2.40–2.31 (m, 2 H, CH_2), 2.16–2.10 (m, 2 H, CH_2), 2.05 (s, 3 H, CH_3), 1.92 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.6, 143.8, 136.3, 134.7, 131.2, 130.4, 129.3, 128.3, 127.5, 127.1, 126.2, 113.1, 39.1, 23.7, 22.7, 17.3, 10.6.

MS (ESI): *m*/*z* (%) = 316.17 (100) $[M + H]^+$.

Anal. Calcd for $C_{22}H_{21}NO$: C, 83.78; H, 6.71; N, 4.44. Found: C, 83.62; H, 6.93; N, 4.18.

3-(4-Methoxyphenyl)-2-methyl-1-(*o*-tolyl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9ii)
Yellow solid; yield: 157.4 mg (91%); mp 122–124 °C.

IR (KBr): 2955, 1720, 1651, 1446, 1361, 1288, 1205, 1121, 1066, 969, 921, 739 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.42–7.34 (m, 5 H, H_{Ar}), 7.24 (d, *J* = 7.6 Hz, 1 H, H_{Ar}), 6.94 (d, *J* = 8.8 Hz, 2 H, H_{Ar}), 3.85 (s, 3 H, OCH_3), 2.54–2.47 (m, 3 H, CH_2), 2.39–2.32 (m, 1 H, CH_2), 2.13–2.05 (m, 5 H, CH_2, CH_3), 1.90 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.7, 158.0, 143.6, 136.4, 136.3, 131.4, 131.1, 129.3, 128.3, 127.7, 127.1, 119.8, 117.4, 113.1, 55.2, 39.1, 23.7, 22.7, 17.3, 10.6.

HRMS (ESI): *m*/*z* $[M + H]^+$ calcd for $C_{23}H_{24}NO_2$: 346.1802; found: 346.1806.

1-(2,4-Dimethylphenyl)-2-methyl-3-(4-nitrophenyl)-1,5,6,7-tetrahydro-4*H*-indol-4-one (9jj)
Yellow solid; yield: 157.6 mg (84%); mp 134–136 °C.

IR (KBr): 2959, 1718, 1654, 1438, 1355, 1321, 1200, 1109, 1074, 970, 901, 729 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.24–8.21 (m, 2 H, H_{Ar}), 7.60 (d, *J* = 8.8 Hz, 2 H, H_{Ar}), 7.22 (s, 1 H, H_{Ar}), 7.17 (d, *J* = 8.0 Hz, 1 H, H_{Ar}), 7.10 (d, *J* = 8.0 Hz, 1 H, H_{Ar}), 2.53–2.47 (m, 3 H, CH_2), 2.43–2.34 (m, 4 H, CH_2, CH_3), 2.13–2.10 (m, 2 H, CH_2), 2.01 (s, 3 H, CH_3), 1.93 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.9, 145.9, 145.1, 142.3, 139.7, 135.7, 133.1, 132.0, 131.0, 129.6, 128.0, 127.9, 127.5, 124.0, 122.9, 118.4, 117.1, 39.0, 23.5, 22.6, 21.2, 17.2, 10.7.

MS (ESI): *m*/*z* (%) = 375.24 (100) $[M + H]^+$, 274.67, 216.28.

Anal. Calcd for $C_{23}H_{22}N_2O_3$: C, 73.78; H, 5.92; N, 7.48. Found: C, 73.85; H, 5.78; N, 7.56.

1-(2,4-Dimethylphenyl)-2-methyl-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9kk)
Yellow solid; yield: 143.6 mg (87%); mp 128–130 °C.

IR (KBr): 2954, 1717, 1649, 1440, 1366, 1331, 1287, 1209, 1103, 974, 921, 727 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.47–7.45 (m, 2 H, H_{Ar}), 7.40–7.36 (m, 2 H, H_{Ar}), 7.29–7.28 (m, 1 H, H_{Ar}), 7.20 (s, 1 H, H_{Ar}), 7.16–

7.10 (m, 2 H, H_{Ar}), 2.54–2.48 (m, 3 H, CH_2), 2.42 (s, 3 H, CH_3), 2.39–2.32 (m, 1 H, CH_2), 2.11–2.06 (m, 2 H, CH_2), 2.01 (s, 3 H, CH_3), 1.91 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.6, 144.0, 139.3, 135.9, 134.8, 133.7, 131.8, 130.4, 128.2, 128.0, 127.8, 127.5, 126.1, 120.1, 117.3, 39.1, 23.7, 22.7, 21.2, 17.2, 10.6.

MS (ESI): *m*/*z* (%) = 330.29 (100) $[M + H]^+$, 274.39, 216.22.

Anal. Calcd for $C_{23}H_{23}NO$: C, 83.85; H, 7.04; N, 4.25. Found: C, 84.12; H, 7.11; N, 4.38.

1-(2-Methoxyphenyl)-2-methyl-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9ll)
Yellow solid; yield: 152.6 mg (92%); mp 107–109 °C.

IR (KBr): 2958, 1719, 1655, 1451, 1366, 1327, 1200, 1070, 966, 745 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.49–7.45 (m, 3 H, H_{Ar}), 7.40–7.36 (m, 2 H, H_{Ar}), 7.26–7.23 (m, 2 H, H_{Ar}), 7.11–7.08 (m, 2 H, H_{Ar}), 3.85 (s, 3 H, OCH_3), 2.55–2.48 (m, 4 H, CH_2), 2.13–2.08 (m, 2 H, CH_2), 1.96 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.7, 155.4, 144.8, 134.9, 130.5, 130.4, 129.6, 128.9, 127.5, 126.1, 125.7, 120.9, 117.4, 112.1, 55.7, 39.1, 23.5, 22.6, 10.5.

HRMS (ESI): *m*/*z* $[M + H]^+$ calcd for $C_{22}H_{22}NO_2$: 332.1645; found: 332.1640.

2-Methyl-1-(naphthalen-1-yl)-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9mm)
Red solid; yield: 135.4 mg (77%); mp 127–129 °C.

IR (KBr): 2957, 1721, 1655, 1439, 1366, 1331, 1228, 1062, 980, 917, 684 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.01–7.99 (m, 2 H, H_{Ar}), 7.65–7.50 (m, 6 H, H_{Ar}), 7.43–7.39 (m, 2 H, H_{Ar}), 7.33–7.29 (m, 2 H, H_{Ar}), 2.54–2.50 (m, 3 H, CH_2), 2.36–2.27 (m, 1 H, CH_2), 2.10–2.04 (m, 2 H, CH_2), 1.90 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.7, 145.1, 134.7, 134.3, 133.9, 130.9, 130.5, 129.6, 129.5, 128.4, 127.9, 127.6, 127.0, 126.3, 126.0, 125.4, 122.5, 120.3, 117.6, 39.1, 23.6, 22.6, 10.5.

MS (ESI): *m*/*z* (%) = 352.23 (100) $[M + H]^+$.

Anal. Calcd for $C_{25}H_{21}NO$: C, 85.44; H, 6.02; N, 3.99. Found: C, 85.18; H, 6.31; N, 3.76.

1-Cyclohexyl-2,6,6-trimethyl-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9nn)
Yellow oil; yield: 139.0 mg (83%).

IR (KBr): 2951, 1729, 1635, 1361, 1337, 1222, 1060, 919, 681 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.36–7.33 (m, 4 H, H_{Ar}), 7.25–7.23 (m, 1 H, H_{Ar}), 4.05–3.97 (m, 1 H, CH), 2.79 (s, 2 H, CH_2), 2.31 (s, 2 H, CH_2), 2.21 (s, 3 H, CH_3), 1.97–1.95 (m, 6 H, CH_2), 1.81–1.78 (m, 1 H, CH_2), 1.48–1.38 (m, 2 H, CH_2), 1.29–1.26 (m, 1 H, CH_2), 1.16 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.7, 141.2, 135.2, 131.8, 130.7, 127.5, 127.4, 126.0, 111.5, 57.0, 52.6, 35.1, 32.2, 29.7, 28.7, 25.5, 25.4.

MS (ESI): *m*/*z* (%) = 335.24 (100) $[M + H]^+$.

Anal. Calcd for $C_{23}H_{29}NO$: C, 82.34; H, 8.71; N, 4.18. Found: C, 82.57; H, 8.49; N, 4.32.

1-Benzyl-2,6,6-trimethyl-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9oo)
Yellow solid; yield: 149.2 mg (87%); mp 156–158 °C.

IR (KBr): 2955, 1723, 1636, 1355, 1329, 1220, 1057, 959, 688 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.43–7.26 (m, 8 H, H_{Ar}), 6.98 (d, *J* = 7.6 Hz, 2 H, H_{Ar}), 5.11 (s, 2 H, CH_2), 2.59 (s, 2 H, CH_2), 2.37 (s, 2 H, CH_2), 2.12 (s, 3 H, CH_3), 1.11 (s, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.8, 142.3, 136.7, 134.7, 130.5, 129.0, 127.9, 127.7, 127.5, 126.1, 125.6, 120.6, 116.3, 53.0, 47.2, 36.3, 35.1, 28.6, 10.2.

MS (ESI): *m*/*z* (%) = 343.16 (100) $[M + H]^+$.

Anal. Calcd for $C_{24}H_{25}NO$: C, 83.93; H, 7.34; N, 4.08. Found: C, 83.79; H, 7.29; N, 4.17.

2,6,6-Trimethyl-1-pentyl-3-phenyl-1,5,6,7-tetrahydro-4*H*-indol-4-one (9pp)
Yellow oil; yield: 137.2 mg (85%).

IR (KBr): 2958, 1722, 1638, 1351, 1322, 1231, 1055, 931, 667 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.39–7.33 (m, 4 H, H_{Ar}), 7.27–7.23 (m, 1 H, H_{Ar}), 3.81 (t, *J* = 7.6 Hz, 2 H, CH_2), 2.65 (s, 2 H, CH_2), 2.34 (s, 2 H, CH_2), 2.21 (s, 3 H, CH_3), 1.71–1.66 (m, 2 H, CH_2), 1.40–1.37 (m, 4 H, CH_2), 1.16 (s, 6 H, CH_3), 0.96 (t, *J* = 7.2 Hz, 3 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.6, 141.6, 134.9, 130.5, 127.5, 127.3, 126.0, 120.3, 115.9, 52.9, 44.2, 36.5, 35.0, 30.5, 29.0, 28.7, 22.4, 14.0, 10.2.

MS (ESI): *m*/*z* (%) = 323.29 (100) $[M + H]^+$.

Anal. Calcd for $C_{22}H_{29}NO$: C, 81.69; H, 9.04; N, 4.33. Found: C, 81.36; H, 9.33; N, 4.18.

5,5-Dimethyl-2-(2-nitro-1-phenylethyl)-3-(phenylamino)cyclohex-2-en-1-one (10)
In a 10-mL reaction vial, nitroolefin (0.5 mmol), 5,5-dimethylcyclohexane-1,3-dione (**1c**, 0.5 mmol), aniline (0.5 mmol), L-proline (0.05 mmol), and H_2O (3.0 mL) were mixed and then capped. The mixture was stirred for 12 h at 45 °C. Upon completion as shown by TLC monitoring, the mixture was cooled to r.t. and extracted with EtOAc (3 × 5 mL). The resulting residue was purified by column chromatography (silica gel, EtOAc–petroleum ether, 1:10) to give the pure product; yellow oil; yield: 125.6 mg (69%).

IR (KBr): 2949, 1710, 1637, 1447, 1356, 1288, 1213, 1106, 981, 929, 718 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.39–7.22 (m, 7 H, H_{Ar}), 7.24 (t, *J* = 7.2 Hz, 1 H, H_{Ar}), 6.89 (d, *J* = 7.6 Hz, 2 H, H_{Ar}), 6.78 (s, 1 H, NH), 5.33–5.28 (m, 1 H, CH_2), 5.20–5.15 (m, 1 H, CH_2), 5.00–4.96 (m, 1 H, CH_2), 2.38–2.24 (m, 4 H, CH_3), 0.99 (d, *J* = 10.4 Hz, 6 H, CH_3).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.4, 159.0, 139.4, 138.1, 129.4, 129.6, 127.4, 127.3, 126.1, 125.1, 109.6, 50.9, 40.9, 39.6, 32.6, 28.8, 27.8.

MS (ESI): *m*/*z* (%) = 365.23 (100) $[M + H]^+$.

Anal. Calcd for $C_{22}H_{24}N_2O_3$: C, 72.51; H, 6.64; N, 7.69. Found: C, 72.39; H, 6.80; N, 7.74.

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Supporting Information for this article is available online at http://www.thieme-connect.com/ejournals/toc/synthesis.

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