Facile Synthesis of Spiro[indane-2,1′-pyrrolo[2,1-*a*]isoquinolines] via Three-Component Reaction of Isoquinolinium Salts, Indane-1,3-dione, and Isatins

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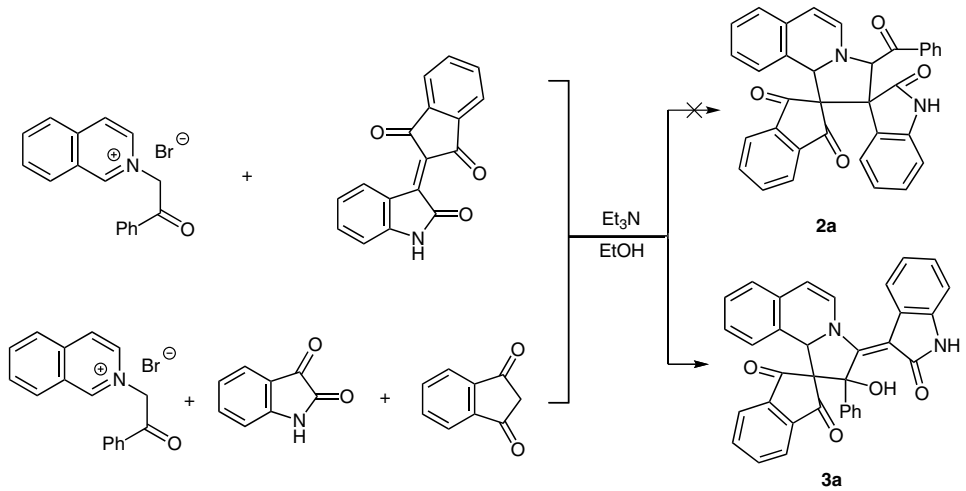
Abstract: A series of novel 2′-aryl-2′-(2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′,3′-dihydro-10b′*H*-spiro[indene-2,1′-pyrrolo[2,1-*a*]isoquinoline]-1,3-diones were efficiently synthesized by three-component reactions of in situ generated *N*-phenacylisoquinolinium bromides with indane-1,3-dione and isatins in ethanol with triethylamine as the base. A domino reaction mechanism for the formation of the spiro compounds was rationally proposed.

Key words: spiro compound, isoquinoline, isatin, indane-1,3-dione, nitrogen ylide, 1,3-cycloaddition

Heteroaromatic N-ylides such as pyridinium, quinolinium, and isoquinolinium methylides are readily available from the alkylations of aza-aromatic heterocycles and sequential deprotonation reactions.[1,2] The heteroaromatic N-ylides have been extensively used in cycloadditions for the synthesis of various nitrogen-containing heterocycles.[3–9] In the past few years, we have successfully developed several multicomponent reactions based on in situ formed reactive pyridinium salts and isoquinolinium salts for the efficient synthesis of versatile heterocyclic compounds.[10,11] Recently, we envisioned that the 1,3-dipolar cycloaddition reaction of heterocyclic N-ylides with isatin derivatives or 3-methyleneoxindoles would provide convenient methods for the construction of important spirooxindole skeletons,[12,13] which are the core structures of a large family of natural alkaloids and many pharmacological agents with important bioactivity and interesting structural properties.[14,15] In this practical field, we found that the reactions of *N*-benzylbenzimidazolium salts, isatin, and malononitrile gave a series of novel zwitterionic salts.[16] The 1,3-cycloaddition reactions of 3-phenacylideneoxindoles with aza-aromatic N-ylides afforded a series of polycyclic spirooxindole systems.[17] On the other hand, similar cycloaddition reactions of pyridinium salts and 4-(dimethylamino)pyridinium salts with 3-phenacylideneoxindoles produced functionalized spiro[cyclopropane-1,3′-indolines], and 3-furan-3(2*H*)-ylideneindolin-2-ones depending upon the structures of the pyridinium salts and reaction conditions.[18] Thus, the very interesting molecular diversity of the cycloaddition of heteroaromatic N-ylides with isatin derivatives so far has prompted us to continue our investigation using the reactivity of other substrates in these reactions. Here we wish to report the unusual formation of novel 2′-isatylidenespiro[indane-2,1′-pyrrolo[2,1-*a*]isoquinolines] [2′-aryl-2′-(2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′,3′-dihydro-10b′*H*-spiro[indene-2,1′-pyrrolo[2,1-*a*]isoquinoline]-1,3-diones] from the three-component reaction of isoquinolinium salts, isatins, and indane-1,3-dione.



Scheme 1 Synthesis of the spiro compound **3a** via multicomponent reaction

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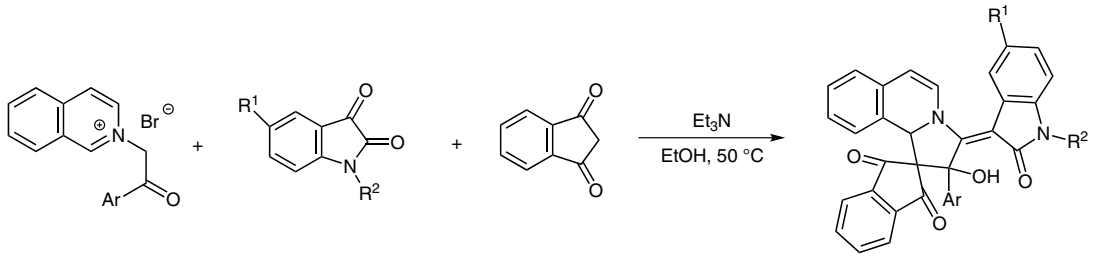
We began our investigation by treating *N*-phenacylisoquinolinium bromide (**1a**) with previously prepared 2-isatylideneindane-1,3-dione[19] in ethanol in the presence of triethylamine according to the previously established reaction conditions for the cycloaddition reactions used for the efficient synthesis of functionalized spiro[indoline-3,1′-pyrrolo[2,1-*a*]isoquinolines][17] (Scheme 1). After workup, we were surprised to find that the expected 1,3-dipolar cycloadduct **2a** was not obtained. Instead, a new spiro compound **3a** was prepared in moderate yield, in which the moieties of isatin and indane-1,3-dione were not connected to each other. This result clearly indicated that a retro-aldol condensation of isatylideneindane-1,3-dione gave the starting substrates, indane-1,3-dione and isatin. Thus, the three-component reaction of *N*-phenacylisoquinolinium bromide (**1a**), isatin, and indane-1,3-dione was further investigated. An ethanol solution of isatin, indane-1,3-dione, and in situ generated *N*-phenacylisoquinolinium bromide (**1a**), from the reaction of isoquinoline and phenacyl bromide in the presence of triethylamine, was heated at ca. 50 °C overnight. The functionalized 2′-isatylidenespiro[indane-2,1′-pyrrolo[2,1-*a*]isoquinoline] **3a** was obtained in 85% yield (Table 1, entry 1). Then under similar conditions, various isatins with 5-methyl, 5-fluoro, and 5-chloro groups and 1-benzyl and 1-butyl groups reacted smoothly with *N*-phenacylisoquinolinium bromide (**1a**) and *N*-(4-chlorophenacyl)isoquinolinium bromide (**1b**) to afford a molecular library of spiro[indane-2,1′-pyrrolo[2,1-*a*]isoquinolines] **3b**–**q** in satisfactory yields (entries 2–17, 72–88%). These results clearly showed that the substituents in isatins showed very little effect on the yields of products and this new cycloaddition of isoquinolinium salts has a wide substrate scope.

Table 1 Synthesis of Spiro[indane-2,1′-pyrrolo[2,1-*a*]isoquinolines][a]



Entry	Product	Ar	R^1	R^2	Yield (%)
1	**3a**	Ph	H	H	85
2	**3b**	Ph	Me	H	86
3	**3c**	Ph	F	H	88
4	**3d**	Ph	Cl	H	87
5	**3e**	4-ClC_6H_4	H	H	82
6	**3f**	4-ClC_6H_4	Me	H	81
7	**3g**	4-ClC_6H_4	F	H	85
8	**3h**	4-ClC_6H_4	Cl	H	84
9	**3i**	Ph	H	Bn	75
10	**3j**	Ph	Me	Bn	76
11	**3k**	Ph	Cl	Bn	74
12	**3l**	Ph	F	Bn	73
13	**3m**	4-ClC_6H_4	F	Bn	72
14	**3n**	Ph	F	Bu	78
15	**3o**	Ph	Me	Bu	80
16	**3p**	Ph	Cl	Bu	80
17	**3q**	4-ClC_6H_4	F	Bu	73

[a] Reaction conditions: (a) 1. isoquinoline (1.2 mmol), phenacyl bromide (1.0 mmol), EtOH (10.0 mL), r.t., 1 h; 2. isatin (1.0 mmol), indane-1,3-dione (1.0 mmol), Et_3N (0.5 mmol), 50 °C, 12 h.
[b] Isolated yields.

The structures of the spiro compounds **3a**–**q** were characterized by ^{1}H and ^{13}C NMR, HRMS, and IR spectra. A typical structure, spiro compound **3e**, was unambiguously confirmed by single-crystal X-ray diffraction (Figure 1). In the ^{1}H NMR spectra of spiro compounds **3a**–**q**, the CH unit and the hydroxy group in the newly formed pyrrole ring usually display two singlets at ca. δ = 5.70 and 8.70; this indicates that only one stereoisomer predominately exists in each sample of the spiro compounds **3a**–**q**. In the molecular structure of compound **3e**, the newly formed pyrrole ring assumes a slightly puckered envelope conformation; the plane of the oxindole deviates from the pyrrole ring. The phenyl group of the isatylidene moiety and the isoquinoline ring are *cis*, that is the tetrasubstituted alkene is the *E*-isomer. It is clearly observed that the hydrogen atom and hydroxy group are *trans*. At present, we tentatively propose that the prepared spiro compounds **3a**–**q** exist in this configuration based on the ^{1}H NMR data and a single crystal structure.

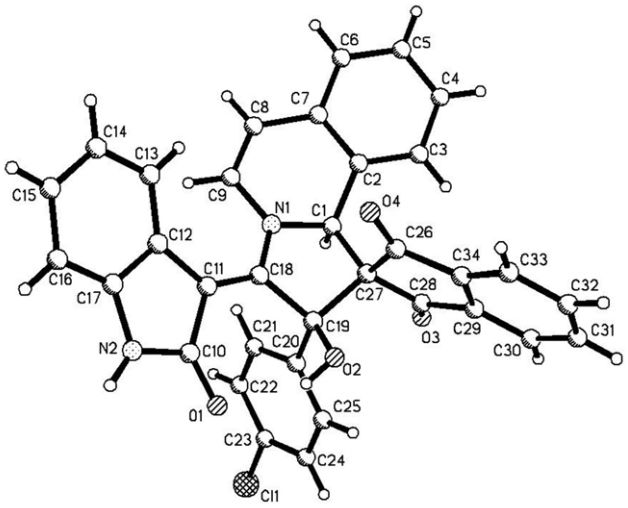
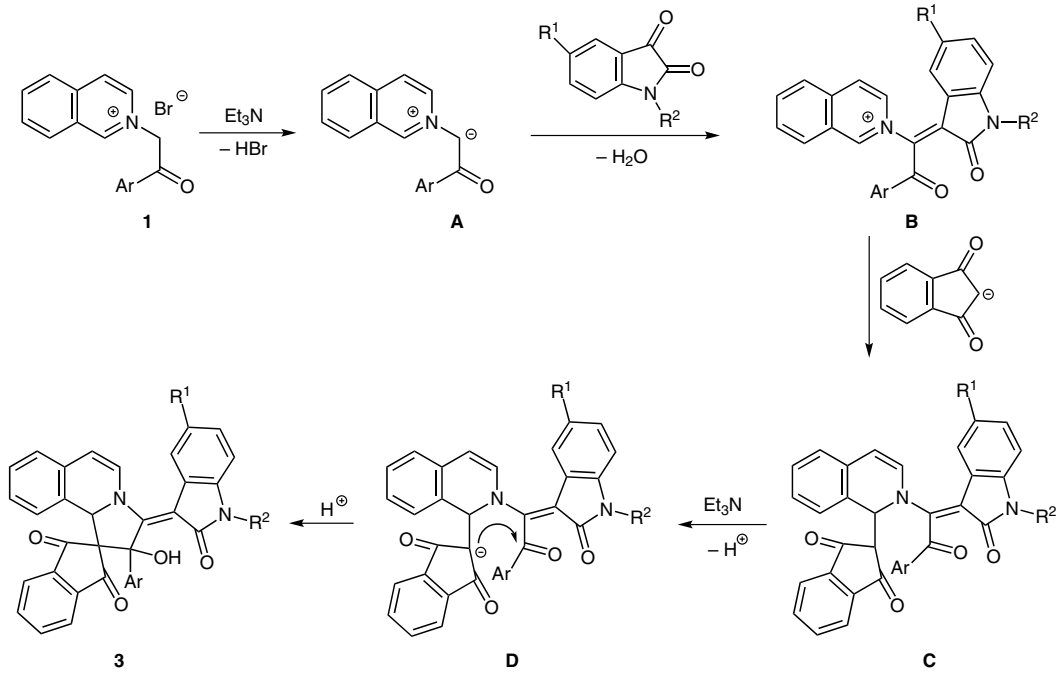


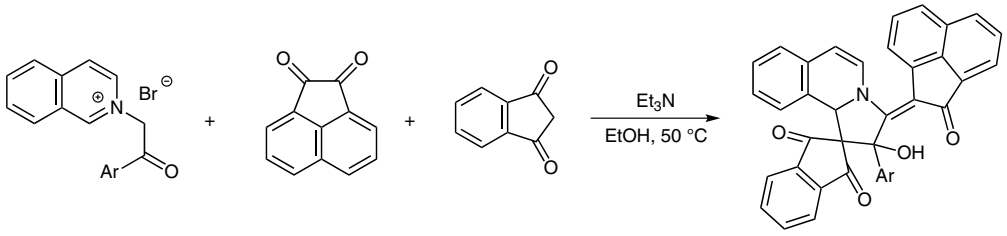
Figure 1 Molecular structure of spiro compound **3e**

To explain the formation of the spiro compounds, a reaction mechanism for this domino three-component reaction is proposed in Scheme 2. Firstly, *N*-phenacylisoquinolinium bromide **1** is deprotonated by triethylamine to give a isoquinolinium ylide **A**. Secondly, the condensation of isoquinolinium ylide **A** with isatin gives isatylideneisoquinolinium salt **B**. Thirdly, the reaction of carbanion of indane-1,3-dione with intermediate **B** affords adduct intermediate **C**, which in turn is deprotonated by triethylamine to give another carbanion **D**. At last, the intramolecular nucleophilic addition of carbanion to carbonyl group in the intermediate **D** and the protonation of hydroxide ion results in the final spiro compound **3**. Because there are no reactive carbonyl groups in the molecules, similar reactions of *N*-(4-nitrobenzyl) or *N*-[(ethoxycarbonyl)methyl]isoquinolinium bromides with isatin and indane-1,3-dione only gave complex mixtures.

In order to explore the potential of this protocol for spiroheterocyclic synthesis, the three-component reaction of isoquinolinium salts, acenaphthenequinone, and indane-1,3-dione was also performed under similar reaction conditions (Scheme 3). The corresponding 2′-acenaphthylidenespiro[indane-2,1′-pyrrolo[2,1-*a*]isoquinolines] **4a**,**b** were obtained in 67% and 72% yields, respectively. This result clearly indicated that this reaction has broad generality.



Scheme 2 The proposed mechanism for the domino reaction



4a Ar = Ph, 67%
4b Ar = 4-ClC$_6$H$_5$, 72%

Scheme 3 Synthesis of 2′-acenaphthylidenespiro[indane-2,1′-pyrrolo[2,1-*a*]isoquinolines]

In summary, we have successfully found an unprecedented domino three-component reaction of isoquinolinium salt, isatins and indane-1,3-dione. The reaction mechanism involved the sequential condensation and cycloaddition reaction of an in situ generated isoquinolinium ylide. This reaction provided a convenient procedure for the synthesis of novel functionalized 2′-isatylidenespiro[indane-2,1′-pyrrolo[2,1-*a*]isoquinolines] in good yields. The short reaction time, readily variable substrates, and easiness of handling render this domino reaction applicable to the synthesis of structurally diverse heterocyclic compounds.

All reactions were monitored by TLC. Melting points were taken on a hot-plate microscope apparatus. IR spectra were obtained on a Bruker Tensor 27 spectrophotometer (KBr disc). NMR spectra were recorded with a Bruker AV-600 spectrometer with DMSO-d_6 as solvent and TMS as internal standard (600 and 150 MHz for ^{1}H and ^{13}C NMR spectra, respectively). HRMS (ESI) were obtained with a Bruker MicroTOF spectrometer. X-ray data were collected on a Bruker Smart APEX-2 CCD diffractometer. Single crystal data for compounds **3e** (CCDC 978440) have been deposited with the Cambridge Crystallographic Data Centre.

2′-(2-Oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′,3′-dihydro-10b′*H*-spiro[indene-2,1′-pyrrolo[2,1-*a*]isoquinoline]-1,3-diones 3a–q via Domino Three-Component Reaction; General Procedure

A mixture of isoquinoline (1.2 mmol) and phenacyl bromide (1.0 mmol) in EtOH (10.0 mL) was stirred at r.t. for ~1 h. Then isatin (1.0 mmol), indane-1,3-dione (1.0 mmol), and Et_3N (0.5 mmol) were added. The mixture was heated at 50 °C for overnight. After cooling, the resulting precipitate was collected by filtration, and this was recrystallized (EtOH–$CHCl_3$) to give the pure product for analysis.

2′-Phenyl-2′-(2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-Substituted 3a

Yellow solid; yield: 0.443 g (85%); mp 202–204 °C.

IR (KBr): 3086, 3006, 2879, 2815, 1750, 1711, 1647, 1543, 1466, 1415, 1347, 1271, 1228, 1186, 1151, 1063, 1002, 974, 858, 785, 750 cm^{-1}.

^{1}H NMR (600 MHz, DMSO-d_6): δ = 10.71 (s, 1 H, NH), 8.63 (s, 1 H, OH), 8.13–8.09 (m, 3 H, H_{Ar}), 8.00–7.98 (m, 1 H, H_{Ar}), 7.46 (d, *J* = 7.8Hz, 1 H, H_{Ar}), 7.39–7.36 (m, 4 H, H_{Ar}), 7.24 (q, *J* = 7.2Hz, 4 H, H_{Ar}), 7.13 (t, *J* = 7.8 Hz, 1 H, H_{Ar}), 7.03 (t, *J* = 7.8 Hz, 1 H, H_{Ar}), 6.96 (t, *J* = 7.8 Hz, 1 H, H_{Ar}), 6.89 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 6.60 (d, *J* = 7.8 Hz, 1 H, CH), 6.10 (d, *J* = 7.2 Hz, 1 H, CH), 5.62 (s, 1 H, CH).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.5, 195.0, 170.9, 159.3, 143.3, 141.9, 138.6, 138.1, 137.2, 136.6, 131.1, 129.2, 128.5, 128.3, 127.9, 126.4, 126.0, 125.2, 123.6, 122.9, 122.7, 122.0, 121.8, 120.8, 113.3, 109.6, 98.0, 88.5, 68.9, 63.3.

HRMS (ESI): *m*/*z* [M + Na]$^+$ calcd for $C_{34}H_{22}N_2NaO_4$: 545.1472; found: 545.1471.

2′-(5-Methyl-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-phenyl-Substituted 3b

Yellow solid; yield: 0.461 g (86%); mp 190–192 °C.

IR (KBr): 3136, 3027, 2965, 1745, 1710, 1650, 1620, 1593, 1556, 1487, 1459, 1426, 1326, 1273, 1235, 1216, 1188, 1149, 1064, 1000, 872, 816, 790, 750 cm^{-1}.

^{1}H NMR (600 MHz, DMSO-d_6): δ = 10.60 (s, 1 H, NH), 8.73 (s, 1 H, OH), 8.11–8.09 (m, 3 H, H_{Ar}), 7.99–7.98 (m, 1 H, H_{Ar}), 7.39–7.35 (m, 4 H, H_{Ar}), 7.29 (s, 1 H, H_{Ar}), 7.25–7.22 (m, 3 H, H_{Ar}), 7.20 (d, *J* = 7.2 Hz, 1 H, H_{Ar}), 6.97–6.93 (m, 2 H, H_{Ar}), 6.78 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 6.58 (d, *J* = 7.8 Hz, 1 H, CH), 6.10 (d, *J* = 7.2 Hz, 1 H, CH), 5.63 (s, 1 H, CH), 2.33 (s, 3 H, CH_3).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.5, 195.0, 170.9, 159.1, 143.3, 141.9, 138.4, 138.3, 137.2, 136.6, 136.4, 131.2, 129.3, 129.1, 128.8, 128.7, 128.5, 128.4, 128.3, 127.8, 126.4, 126.0, 125.8, 123.6, 122.9, 122.8, 122.6, 122.0, 112.7, 109.3, 98.2, 88.4, 69.0, 63.2, 21.3.

HRMS (ESI): *m*/*z* [M + Na]$^+$ calcd for $C_{35}H_{24}N_2NaO_4$: 559.1628; found: 559.1624.

2′-(5-Fluoro-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-phenyl-Substituted 3c

Yellow solid; yield: 0.476 g (88%); mp 206–208 °C.

IR (KBr): 3082, 2939, 2842, 1749, 1711, 1648, 1540, 1465, 1409, 1336, 1270, 1238, 1190, 1148, 1063, 1003, 960, 858, 819, 785 cm^{-1}.

^{1}H NMR (600 MHz, DMSO-d_6): δ = 10.74 (s, 1 H, NH), 8.58 (s, 1 H, OH), 8.13–8.09 (m, 3 H, H_{Ar}), 7.99–7.93 (m, 1 H, H_{Ar}), 7.40–7.36 (m, 4 H, H_{Ar}), 7.26–7.23 (m, 4 H, H_{Ar}), 7.19–7.17 (m, 1 H, H_{Ar}), 6.99–6.93 (m, 2 H, H_{Ar}), 6.88–6.85 (m, 1 H, H_{Ar}), 6.67 (d, *J* = 7.8 Hz, 1 H, CH), 6.11 (d, *J* = 7.8 Hz, 1 H, CH), 5.68 (s, 1 H, CH).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.4, 194.9, 171.0, 160.4, 158.1, 156.5, 143.3, 142.0, 138.0, 137.2, 136.7, 134.9, 130.9, 129.3, 128.3, 128.1, 126.6, 126.0, 124.0, 123.6, 122.9, 122.1, 114.1, 111.3, 111.1, 110.1, 108.8, 108.6, 97.7, 88.5, 69.1, 63.4.

HRMS (ESI): *m*/*z* [M + Na]$^+$ calcd for $C_{34}H_{21}FN_2NaO_4$: 563.1378; found: 563.1379.

2′-(5-Chloro-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-phenyl-Substituted 3d

Yellow solid; yield: 0.484 g (87%); mp 214–216 °C.

IR (KBr): 3130, 3062, 2836, 1746, 1710, 1649, 1548, 1461, 1334, 1303, 1272, 1228, 1186, 1147, 1062, 1001, 817, 792, 744 cm^{-1}.

^{1}H NMR (600 MHz, DMSO-d_6): δ = 10.84 (s, 1 H, NH), 8.48 (s, 1 H, OH), 8.13–8.09 (m, 3 H, H_{Ar}), 8.00–7.98 (m, 1 H, H_{Ar}), 7.41–7.37 (m, 5 H, H_{Ar}), 7.26–7.23 (m, 4 H, H_{Ar}), 7.16–7.14 (m, 1 H,

H_{Ar}), 6.98 (t, *J* = 7.6 Hz, 1 H, H_{Ar}), 6.89 (d, *J* = 8.4 Hz, 1 H, H_{Ar}), 6.67 (d, *J* = 7.2 Hz, 1 H, CH), 6.12 (d, *J* = 7.8 Hz, 1 H, CH), 5.70 (s, 1 H, CH).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.5, 194.8, 170.7, 160.4, 143.2, 142.0, 137.8, 137.3, 137.2, 136.7, 130.8, 129.2, 128.6, 128.4, 128.2, 128.1, 126.7, 125.9, 124.7, 124.5, 123.7, 122.9, 122.1, 121.2, 114.0, 110.7, 96.9, 88.5, 69.1, 63.4.

HRMS (ESI): *m*/*z* [M + Na]$^+$ calcd for $C_{34}H_{21}ClN_2NaO_4$: 579.1082; found: 579.1082.

2′-(4-Chlorophenyl)-2′-(2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-Substituted 3e

Yellow solid; yield: 0.456 g (82%); mp 191–192 °C.

IR (KBr): 3090, 2818, 1745, 1709, 1653, 1555, 1467, 1421, 1346, 1272, 1229, 1188, 1149, 1093, 999, 790, 750 cm^{-1}.

^{1}H NMR (600 MHz, DMSO-d_6): δ = 10.78 (s, 1 H, NH), 8.69 (s, 1 H, OH), 8.14–8.10 (m, 3 H, H_{Ar}), 8.00–7.99 (m, 1 H, H_{Ar}), 7.46–7.44 (m, 3 H, H_{Ar}), 7.36 (d, *J* = 7.2 Hz, 1 H, H_{Ar}), 7.28–7.22 (m, 4 H, H_{Ar}), 7.12 (t, *J* = 7.8 Hz, 1 H, H_{Ar}), 7.03 (t, *J* = 7.8 Hz, 1 H, H_{Ar}), 6.97 (t, *J* = 7.8 Hz, 1 H, H_{Ar}), 6.90 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 6.60 (d, *J* = 7.8 Hz, 1 H, CH), 6.10 (d, *J* = 7.8 Hz, 1 H, CH), 5.64 (s, 1 H, CH).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.2, 195.1, 170.9, 158.7, 143.3, 141.9, 138.7, 127.4, 136.7, 133.2, 131.0, 129.1, 128.5, 128.3, 128.0, 127.9, 126.4, 125.3, 123.7, 122.9, 122.6, 122.1, 121.9, 113.3, 109.7, 98.0, 87.9, 68.8, 63.3.

HRMS (ESI): *m*/*z* [M + Na]$^+$ calcd for $C_{34}H_{21}ClN_2NaO_4$: 579.1082; found: 579.1081.

2′-(4-Chlorophenyl)-2′-(5-methyl-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-Substituted 3f

Yellow solid; yield: 0.462 g (81%); mp 220–222 °C.

IR (KBr): 3134, 3025, 1742, 1708, 1684, 1652, 1556, 1488, 1460, 1428, 1338, 1272, 1235, 1217, 1186, 1148, 1092, 1001, 965, 872, 812, 789, 751 cm^{-1}.

^{1}H NMR (600 MHz, DMSO-d_6): δ = 10.66 (s, 1 H, NH), 8.80 (s, 1 H, OH), 8.13–8.09 (m, 3 H, H_{Ar}), 7.99–7.98 (m, 1 H, H_{Ar}), 7.44 (d, *J* = 8.4 Hz, 2 H, H_{Ar}), 7.37 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 7.29 (s, 1 H, H_{Ar}), 7.26–7.23 (m, 3 H, H_{Ar}), 7.19 (d, *J* = 7.2 Hz, 1 H, H_{Ar}), 6.97–6.93 (m, 2 H, H_{Ar}), 6.78 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 6.58 (d, *J* = 7.8 Hz, 1 H, CH), 6.10 (d, *J* = 7.2 Hz, 1 H, CH), 5.65 (s, 1 H, CH), 2.33 (s, 3 H, CH_3).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.2, 195.1, 170.9, 158.4, 143.3, 141.8, 137.5, 137.4, 136.4, 133.1, 131.1, 129.4, 128.9, 128.5, 128.3, 127.9, 126.5, 125.9, 123.7, 122.9, 122.7, 122.1, 112.7, 109.4, 98.2, 87.8, 68.8, 63.2, 21.3.

HRMS (ESI): *m*/*z* [M + Na]$^+$ calcd for $C_{35}H_{23}ClN_2NaO_4$: 593.1239; found: 593.1231.

2′-(4-Chlorophenyl)-2′-(5-fluoro-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-Substituted 3g

Yellow solid; yield: 0.488 g (85%); mp 211–213 °C.

IR (KBr): 3087, 1744, 1708, 1654, 1556, 1465, 1333, 1274, 1240, 1187, 1149, 1093, 1002, 966, 813, 789, 750 cm^{-1}.

^{1}H NMR (600 MHz, DMSO-d_6): δ = 10.81 (s, 1 H, NH), 8.66 (s, 1 H, OH), 8.13–8.11 (m, 3 H, H_{Ar}), 7.99 (s, 1 H, H_{Ar}), 7.45 (d, *J* = 8.4 Hz, 2 H, H_{Ar}), 7.40 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 7.27–7.23 (m, 4 H, H_{Ar}), 7.18 (d, *J* = 10.2 Hz, 1 H, H_{Ar}), 6.99–6.94 (m, 2 H, H_{Ar}), 6.88–6.86 (m, 1 H, H_{Ar}), 6.67 (d, *J* = 7.2 Hz, 1 H, CH), 6.11 (d, *J* = 7.2 Hz, 1 H, CH), 5.70 (s, 1 H, CH).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.1, 194.9, 170.9, 159.7, 158.1, 156.5, 143.3, 141.9, 137.4, 137.1, 136.7, 134.8, 133.3, 130.7, 129.1, 128.6, 128.3, 128.1, 127.9, 126.7, 123.7, 123.8, 123.7, 123.0, 122.1, 114.1, 111.4, 111.2, 110.1, 108.8, 108.6, 97.7, 87.9, 68.9, 63.4.

HRMS (ESI): *m*/*z* [M + Na]$^+$ calcd for $C_{34}H_{20}FClN_2NaO_4$: 597.0988; found: 597.0982.

2′-(5-Chloro-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-(4-chlorophenyl)-Substituted 3h

Yellow solid; yield: 0.496 g (84%); mp 214–216 °C.

IR (KBr): 3126, 2995, 2837, 1743, 1708, 1654, 1554, 1464, 1433, 1401, 1333, 1303, 1271, 1229, 1187, 1149, 1093, 1001, 963, 872, 794, 751 cm^{-1}.

^{1}H NMR (600 MHz, DMSO-d_6): δ = 10.88 (s, 1 H, NH), 8.57 (s, 1 H, OH), 8.14–8.10 (m, 3 H, H_{Ar}), 7.99–7.98 (m, 1 H, H_{Ar}), 7.45 (d, *J* = 9.0 Hz, 2 H, H_{Ar}), 7.40 (d, *J* = 7.8 Hz, 2 H, H_{Ar}), 7.27–7.24 (m, 3 H, H_{Ar}), 7.21 (d, *J* = 7.2 Hz, 1 H, H_{Ar}), 7.16–7.15 (m, 1 H, H_{Ar}), 6.98 (t, *J* = 7.8 Hz, 1 H, H_{Ar}), 6.90 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 6.66 (d, *J* = 7.8 Hz, 1 H, CH), 6.12 (d, *J* = 7.8 Hz, 1 H, CH), 5.73 (s, 1 H, CH).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.3, 194.9, 170.5, 159.7, 143.2, 141.8, 137.5, 137.0, 136.9, 136.8, 133.3, 130.7, 129.0, 128.7, 128.4, 128.2, 127.9, 127.8, 126.8, 124.8, 124.6, 124.3, 123.8, 123.0, 122.0, 121.2, 114.1, 110.8, 96.8, 87.8, 68.8, 63.4.

HRMS (ESI): *m*/*z* [M + Na]$^+$ calcd for $C_{34}H_{20}Cl_2N_2NaO_4$: 613.0692; found: 613.0681.

2′-(1-Benzyl-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-phenyl-Substituted 3i

Yellow solid; yield: 0.459 g (75%); mp 212–214 °C.

IR (KBr): 3062, 2920, 1747, 1707, 1635, 1603, 1536, 1479, 1421, 1361, 1269, 1182, 1152, 1077, 1011, 976, 799 cm^{-1}.

^{1}H NMR (600 MHz, DMSO-d_6): δ = 8.15–8.10 (m, 3 H, H_{Ar}), 8.00 (br s, 1 H, OH), 7.51 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 7.41–7.36 (m, 4 H, H_{Ar}), 7.30–7.24 (m, 7 H, H_{Ar}), 7.21–7.17 (m, 3 H, H_{Ar}), 7.13 (t, *J* = 7.2 Hz, 1 H, H_{Ar}), 7.07 (t, *J* = 7.2 Hz, 1 H, H_{Ar}), 7.03 (d, *J* = 7.2 Hz, 1 H, H_{Ar}), 6.97 (t, *J* = 7.2 Hz, 1 H, H_{Ar}), 6.62 (d, *J* = 7.8 Hz, 1 H, CH), 6.11 (d, *J* = 7.8 Hz, 1 H, CH), 5.68 (s, 1 H, CH), 4.86 (q, *J* = 15.6 Hz, 2 H, CH_2).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.4, 194.9, 168.8, 159.8, 143.2, 142.0, 138.8, 138.0, 136.7, 131.0, 129.2, 128.5, 128.4, 128.0, 126.5, 125.9, 125.0, 123.6, 123.0, 122.0, 121.9, 121.7, 121.5, 113.7, 109.1, 96.6, 88.6, 69.0, 63.3, 42.7.

HRMS (ESI): *m*/*z* [M + H]$^+$ calcd for $C_{41}H_{29}N_2O_4$: 613.2122; found: 613.2117.

2′-(1-Benzyl-5-methyl-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-phenyl-Substituted 3j

Yellow solid; yield: 0.476 g (76%); mp 218–220 °C.

IR (KBr): 3062, 2915, 1745, 1708, 1646, 1593, 1542, 1488, 1458, 1421, 1383, 1356, 1273, 1185, 1077, 1011, 880, 791 cm^{-1}.

^{1}H NMR (600 MHz, DMSO-d_6): δ = 8.45 (s, 1 H, OH), 8.14–8.09 (m, 3 H, H_{Ar}), 8.01–7.99 (m, 1 H, H_{Ar}), 7.40–7.37 (m, 4 H, H_{Ar}), 7.34 (s, 1 H, H_{Ar}), 7.27–7.23 (m, 6 H, H_{Ar}), 7.20–7.18 (m, 1 H, H_{Ar}), 7.14 (d, *J* = 7.8 Hz, 2 H, H_{Ar}), 6.99–6.94 (m, 2 H, H_{Ar}), 6.90 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 6.61 (d, *J* = 7.8 Hz, 1 H, CH), 6.11 (d, *J* = 7.8 Hz, 1 H, CH), 5.69 (s, 1 H, CH), 4.83 (q, *J* = 16.2 Hz, 2 H, CH_2), 2.33 (s, 3 H, CH_3).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.4, 195.0, 168.9, 159.5, 143.3, 142.0, 138.1, 137.2, 136.7, 131.0, 129.1, 128.4, 128.3, 127.9, 127.2, 126.5, 125.9, 125.6, 123.6, 122.9, 122.5, 122.0, 121.9, 113.1, 108.8, 96.8, 88.5, 69.1, 63.2, 42.7, 21.3.

HRMS (ESI): *m*/*z* [M + H]$^+$ calcd for $C_{42}H_{31}N_2O_4$: 627.2278; found: 627.2275.

2′-(1-Benzyl-5-chloro-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-phenyl-Substituted 3k

Yellow solid; yield: 0.478 g (74%); mp 220–222 °C.

IR (KBr): 1747, 1708, 1646, 1550, 1476, 1351, 1272, 1180, 1081, 1010, 877, 793, 747 cm^{-1}.

^1H NMR (600 MHz, DMSO-d_6): δ = 8.23 (s, 1 H, OH), 8.14–8.10 (m, 3 H, H_{Ar}), 8.01–7.99 (m, 1 H, H_{Ar}), 7.45–7.44 (m, 1 H, H_{Ar}), 7.43–7.39 (m, 4 H, H_{Ar}), 7.29–7.24 (m, 6 H, H_{Ar}), 7.21–7.17 (m, 2 H, H_{Ar}), 7.14 (d, J = 7.2 Hz, 2 H, H_{Ar}), 7.04 (d, J = 8.4 Hz, 1 H, H_{Ar}), 6.99 (t, J = 7.8 Hz, 1 H, H_{Ar}), 6.70 (d, J = 1.2 Hz, 1 H, CH), 6.13 (d, J = 7.2 Hz, 1 H, CH), 5.77 (s, 1 H, CH), 4.86 (q, J = 15.6 Hz, 2 H, CH_2).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.4, 194.8, 168.6, 160.9, 143.3, 142.0, 137.7, 137.3, 136.8, 136.4, 130.7, 129.2, 128.6, 128.5, 128.4, 128.3, 128.0, 127.3, 126.8, 125.7, 125.5, 124.2, 123.7, 123.0, 122.1, 121.1, 110.2, 95.5, 88.6, 69.3, 63.4, 42.7.

HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{41}H_{27}ClN_2NaO_4$: 669.1552; found: 669.1543.

2′-(1-Benzyl-5-fluoro-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-phenyl-Substituted 3l

Yellow solid; yield: 0.460 g (73%); mp 211–213 °C.

IR (KBr): 3064, 1746, 1709, 1639, 1594, 1534, 1483, 1449, 1423, 1356, 1268, 1174, 1079, 1011, 955, 882, 795 cm^{-1}.

^1H NMR (600 MHz, DMSO-d_6): δ = 8.31 (s, 1 H, OH), 8.14–8.10 (m, 3 H, H_{Ar}), 8.00–7.99 (m, 1 H, H_{Ar}), 7.41–7.39 (m, 4 H, H_{Ar}), 7.31–7.27 (m, 4 H, H_{Ar}), 7.25 (d, J = 7.2 Hz, 3 H, H_{Ar}), 7.23–7.21 (m, 1 H, H_{Ar}), 7.16 (d, J = 7.8 Hz, 2 H, H_{Ar}), 7.03–6.99 (m, 3 H, H_{Ar}), 6.70 (d, J = 7.2 Hz, 1 H, CH), 6.13 (d, J = 7.8 Hz, 1 H, CH), 5.74 (s, 1 H, CH), 4.86 (q, J = 15.6 Hz, 2 H, CH_2).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.3, 194.8, 168.8, 160.8, 156.9, 143.3, 142.0, 137.8, 137.3, 136.7, 136.5, 134.9, 130.7, 129.2, 128.6, 128.5, 128.4, 128. 2, 128.0, 127.3, 126.8, 125.9, 123.7, 123.0, 122.1, 114.4, 111.1, 109.6, 108.8, 108.6, 96.2, 88.6, 69.2, 63.4, 42.7.

HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{41}H_{27}FN_2NaO_4$: 653.1847; found: 653.1842.

2′-(1-Benzyl-5-fluoro-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-(4-chlorophenyl)-Substituted 3m

Yellow solid; yield: 0.478 g (72%); mp 224–226 °C.

IR (KBr): 3071, 1747, 1712, 1638, 1537, 1483, 1345, 1269, 1175, 1093, 1010, 955, 885, 797, 750 cm^{-1}.

^1H NMR (600 MHz, DMSO-d_6): δ = 8.41 (s, 1 H, OH), 8.15–8.10 (m, 3 H, H_{Ar}), 8.01–7.99 (m, 1 H, H_{Ar}), 7.46 (d, J = 8.4 Hz, 2 H, H_{Ar}), 7.40 (d, J = 7.2 Hz, 1 H, H_{Ar}), 7.30–7.20 (m, 7 H, H_{Ar}), 7.15 (d, J = 7.2 Hz, 2 H, H_{Ar}), 7.03–6.97 (m, 3 H, H_{Ar}), 6.68 (d, J = 7.2 Hz, 1 H, CH), 6.13 (d, J = 7.2 Hz, 1 H, CH), 5.79 (s, 1 H, CH), 5.77 (s, 1 H, CH), 4.86 (q, J = 15.6 Hz, 2 H, CH_2).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.1, 195.0, 168.8, 160.1, 157.0, 143.3, 141.9, 137.4, 137.0, 136.8, 136.4, 134.9, 133.3, 130.6, 129.1, 128.6, 128.5, 128.4, 128.3, 127.9, 127.8, 127.3, 126.8, 123.8, 123.1, 123.0, 122.1, 114.4, 111.2, 96.3, 88.0, 69.1, 63.4, 56.0. 42.8.

HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{41}H_{26}ClFN_2NaO_4$: 687.1457; found: 687.1450.

2′-(1-Butyl-5-fluoro-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-phenyl-Substituted 3n

Yellow solid; yield: 0.465 g (78%); mp 217–219 °C.

IR (KBr): 3084, 2869, 1749, 1711, 1638, 1541, 1483, 1456, 1382, 1356, 1270, 1186, 1143, 1104, 1004, 953, 874, 796 cm^{-1}.

^1H NMR (600 MHz, DMSO-d_6): δ = 8.35 (s, 1 H, OH), 8.14–8.10 (m, 3 H, H_{Ar}), 8.00–7.99 (m, 1 H, H_{Ar}), 7.42–7.37 (m, 4 H, H_{Ar}), 7.28–7.22 (m, 5 H, H_{Ar}), 7.12–7.10 (m, 1 H, H_{Ar}), 7.06–7.03 (m, 1 H, H_{Ar}), 6.98 (t, J = 7.8 Hz, 1 H, H_{Ar}), 6.70 (d, J = 7.8 Hz, 1 H, CH), 6.11 (d, J = 7.8 Hz, 1 H, CH), 5.69 (s, 1 H, CH), 3.65–3.57 (m, 2 H, CH_2), 1.45–1.40 (m, 2 H, CH_2), 1.19 (q, J = 7.2 Hz, 2 H, CH_2), 0.80 (t, J = 7.2 Hz, 3 H, CH_3).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.4, 194.8, 168.7, 160.5, 158.4, 156.8, 143.3, 142.0, 137.8, 137.2, 136.7, 135.3, 130.8, 129.3, 128.5, 128.4, 128.2, 128.1, 126.7, 125.9, 123.6, 123.1, 123.0, 122.9, 122.1, 114.3, 111.1, 111.0, 109.2, 108.8, 108.6, 96.0, 88.5, 69.2, 63.4, 29.4, 19.5, 13.4.

HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{38}H_{29}FN_2NaO_4$: 619.2004; found: 619.2004.

2′-(1-Butyl-5-methyl-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-phenyl-Substituted 3o

Yellow solid; yield: 0.474 g (80%); mp 210–212 °C.

IR (KBr): 3072, 2866, 1746, 1709, 1640, 1595, 1553, 1490, 1443, 1422, 1385, 1359, 1273, 1205, 1142, 1119, 1075, 999, 877, 792, 750 cm^{-1}.

^1H NMR (600 MHz, DMSO-d_6): δ = 8.49 (s, 1 H, OH), 8.13–8.09 (m, 3 H, H_{Ar}), 8.00–7.98 (m, 1 H, H_{Ar}), 7.38–7.34 (m, 5 H, H_{Ar}), 7.25–7.21 (m, 4 H, H_{Ar}), 7.02–6.95 (m, 3 H, H_{Ar}), 6.61 (d, J = 7.8 Hz, 1 H, CH), 6.10 (d, J = 7.8 Hz, 1 H, CH), 5.64 (s, 1 H, CH), 3.58 (t, J = 7.8 Hz, 2 H, CH_2), 2.35 (s, 3 H, CH_3), 1.46–1.40 (m, 2 H, CH_2), 1.18 (q, J = 7.8 Hz, 2 H, CH_2), 0.80 (t, J = 7.2 Hz, 3 H, CH_3).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.5, 195.0, 168.7, 159.2, 143.2, 141.9, 138.1, 137.3, 137.0, 131.2, 129.9, 128.6, 128.5, 128.3, 127.9, 126.5, 123.6, 122.9, 122.5, 122.0, 121.8, 113.0, 97.1, 88.4, 69.0, 63.2, 29.4, 21.3, 19.5, 13.5.

HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{39}H_{33}N_2O_4$: 593.2435; found: 593.2433.

2′-(1-Butyl-5-chloro-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-phenyl-Substituted 3p

Yellow solid; yield: 0.490 g (80%); mp 218–220 °C.

IR (KBr): 3075, 2929, 2869, 1748, 1710, 1640, 1544, 1477, 1426, 1384, 1354, 1272, 1206, 1142, 1117, 1000, 875, 794, 744 cm^{-1}.

^1H NMR (600 MHz, DMSO-d_6): δ = 8.24 (s, 1 H, OH), 8.13–8.11 (m, 3 H, H_{Ar}), 7.99 (s, 1 H, H_{Ar}), 7.44–7.38 (m, 5 H, H_{Ar}), 7.27–7.24 (m, 5 H, H_{Ar}), 7.14 (d, J = 8.4 Hz, 1 H, H_{Ar}), 7.00–6.98 (m, 1 H, H_{Ar}), 6.69 (d, J = 7.8 Hz, 1 H, CH), 6.12 (d, J = 7.8 Hz, 1 H, CH), 5.71 (s, 1 H, CH), 3.63–3.60 (m, 2 H, CH_2), 1.46–1.40 (m, 2 H, CH_2), 1.18 (q, J = 7.2 Hz, 2 H, CH_2), 0.80 (t, J = 7.2 Hz, 3 H, CH_3).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.4, 194.7, 168.5, 160.6, 143.2, 142.0, 137.7, 137.3, 136.7, 130.8, 129.2, 128.6, 128.4, 128.2, 128.1, 126.7, 125.9, 125.2, 124.4, 123.6, 122.9, 122.1, 121.1, 114.2, 109.9, 95.8, 88.5, 69.2, 63.4, 29.4, 19.5, 13.4.

HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{38}H_{30}ClN_2O_4$: 613.1889; found: 613.1883.

2′-(1-Butyl-5-fluoro-2-oxo-1,2-dihydro-3*H*-indol-3-ylidene)-2′-(4-chlorophenyl)-Substituted 3q

Yellow solid; yield: 0.460 g (73%); mp 222–224 °C.

IR (KBr): 3078, 2953, 2870, 1749, 1712, 1642, 1593, 1547, 1484, 1459, 1424, 1355, 1271, 1185, 1144, 1094, 1003, 954, 875, 795, 750 cm^{-1}.

^1H NMR (600 MHz, DMSO-d_6): δ = 8.43 (s, 1 H, OH), 8.14–8.10 (m, 3 H, H_{Ar}), 7.99–7.98 (m, 1 H, H_{Ar}), 7.45 (d, J = 9.0 Hz, 2 H, H_{Ar}), 7.41 (d, J = 7.8 Hz, 1 H, H_{Ar}), 7.28–7.22 (m, 5 H, H_{Ar}), 7.12–7.10 (m, 1 H, H_{Ar}), 7.06–7.02 (m, 1 H, H_{Ar}), 6.98 (t, J = 7.5 Hz, 1 H, H_{Ar}), 6.67 (d, J = 7.8 Hz, 1 H, CH), 6.12 (d, J = 7.8 Hz, 1 H, CH), 5.73 (s, 1 H, CH), 3.65–3.60 (m, 2 H, CH_2), 1.47–1.41 (m, 2 H, CH_2), 1.21–1.15 (m, 2 H, CH_2), 0.80 (t, J = 7.3 Hz, 3 H, CH_3).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.2, 194.9, 168.6, 159.8, 158.4, 156.8, 143.2, 141.9, 137.4, 137.0, 136.8, 135.3, 133.3, 130.7, 129.1, 128.6, 128.4, 128.2, 128.0, 127.9, 126.8, 123.8, 123.0, 122.9, 122.1, 114.3, 111.3, 111.1, 109.3, 108.7, 96.5, 87.9, 68.9, 63.4, 29.4, 19.5, 13.5.

HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{38}H_{29}ClFN_2O_4$: 631.1794; found: 631.1787.

2′-[2-Oxoacenaphthen-1(2*H*)-ylidene]-2′,3′-dihydro-10b′*H*-spiro[indene-2,1′-pyrrolo[2,1-*a*]isoquinoline]-1,3-diones 4a,b via Domino Three-Component Reaction; General Procedure

A mixture of isoquinoline (1.2 mmol) and phenacyl bromide (1.0 mmol) in EtOH (10.0 mL) was stirred at r.t. for ~1 h. Then acenaphthenequinone (1.0 mmol), indane-1,3-dione (1.0 mmol), and Et_3N (0.5 mmol) were added. The mixture was heated at 50 °C for overnight. After cooling, the resulting precipitate was collected by filtration, and this was recrystallized (EtOH–$CHCl_3$) to give the pure product for analysis.

2′-Phenyl-Substituted 4a

Yellow solid; yield: 0.428 g (67%); mp 214–216 °C.

IR (KBr): 3058, 1745, 1708, 1648, 1618, 1520, 1462, 1414, 1347, 1271, 1224, 1151, 1088, 1014, 974, 871, 774 cm^{-1}.

^1H NMR (600 MHz, DMSO-d_6): δ = 8.19 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 8.14 (br s, 1 H, H_{Ar}), 8.11 (s, 2 H, H_{Ar}), 8.01 (s, 1 H, H_{Ar}), 7.83 (d, *J* = 7.2 Hz, 1 H, H_{Ar}), 7.77 (d, *J* = 5.4 Hz, 1 H, H_{Ar}), 7.71–7.69 (m, 3 H, H_{Ar}), 7.45 (d, *J* = 6.6 Hz, 1 H, H_{Ar}), 7.40 (br s, 4 H, H_{Ar}), 7.35 (br s, 3 H, H_{Ar}), 7.27–7.25 (m, 1 H, H_{Ar}), 7.00–6.98 (m, 1 H, H_{Ar}), 6.65 (d, *J* = 6.0 Hz, 1 H, CH), 6.12 (d, *J* = 6.6 Hz, 1 H, CH), 5.70 (s, 1 H, CH).

^{13}C NMR (150 MHz, DMSO-d_6): δ = 195.4, 194.9, 192.7, 159.6, 143.3, 142.0, 137.8, 137.3, 136.7, 134.9, 134.0, 133.4, 131.8, 131.0, 129.6, 128.7, 128.6, 128.5, 128.4, 128.0, 127.9, 126.5, 126.0, 123.7, 123.1, 123.0, 122.1, 122.0, 120.2, 113.8, 105.7, 88.7, 69.0, 63.4.

HRMS (ESI): *m*/*z* $[M + Na]^+$ calcd for $C_{38}H_{23}NNaO_4$: 580.1519; found: 580.1512.

2′-(4-Chlorophenyl)-Substituted 4b

Yellow solid; yield: 0.425 g (72%); mp 204–206 °C.

IR (KBr): 3068, 2831, 1748, 1711, 1653, 1616, 1541, 1489, 1460, 1417, 1345, 1271, 1224, 1152, 1087, 1011, 970, 932, 872, 828, 778, 750 cm^{-1}.

^1H NMR (600 MHz, DMSO-d_6): δ = 8.20 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 8.17–8.15 (m, 1 H, H_{Ar}), 8.13–8.10 (m, 2 H, H_{Ar}), 8.01–8.0 (m, 1 H, H_{Ar}), 7.85–7.84 (m, 1 H, H_{Ar}), 7.78 (d, *J* = 6.6 Hz, 1 H, H_{Ar}), 7.73 (d, *J* = 7.8 Hz, 1 H, H_{Ar}), 7.71–7.69 (m, 2 H, H_{Ar}), 7.59 (s, 1 H, H_{Ar}), 7.47 (d, *J* = 8.4 Hz, 2 H, H_{Ar}), 7.43 (d, *J* = 7.2 Hz, 1 H, H_{Ar}), 7.40 (d, *J* = 7.2 Hz, 1 H, H_{Ar}), 7.36 (d, *J* = 8.4 Hz, 2 H, H_{Ar}), 7.27 (t, *J* = 7.5 Hz, 1 H, H_{Ar}), 6.99 (t, *J* = 7.6 Hz, 1 H, H_{Ar}), 6.65 (d, *J* = 7.8 Hz, 1 H, CH), 6.13 (d, *J* = 7.8 Hz, 1 H, CH), 5.72 (s, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 195.6, 195.4, 193.2, 159.4, 143.7, 142.3, 137.9, 137.5, 137.3, 135.4, 134.4, 133.8, 132.4, 131.4, 130.1, 129.6, 129.0, 128.9, 128.5, 128.4, 127.1, 124.2, 123.6, 123.5, 122.5, 120.7, 114.3, 106.2, 88.7, 69.3, 63.8.

HRMS (ESI): *m*/*z* $[M + Na]^+$ calcd for $C_{38}H_{22}ClNNaO_4$: 614.1130; found: 614.1125.

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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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