An Improved and Benign Synthesis of 9,10-Diarylacridine-1,8-dione and Indenoquinoline Derivatives from 3-Anilino-5,5-dimethylcyclohex-2-enones, Benzaldehydes, and 1,3-Dicarbonyl Compounds in an Ionic Liquid Medium

Xiang-Shan Wang,*[a,b,c] Mei-Mei Zhang,[a] Hong Jiang,[c] Da-Qing Shi,[a,c] Shu-Jiang Tu,[a,c] Xian-Yong Wei,[b] Zhi-Min Zong[b]

[a] Department of Chemistry, Xuzhou Normal University, Xuzhou Jiangsu, 221116, P. R. of China

[b] School of Chemical Engineering, China University of Mining and Technology, Xuzhou Jiangsu 221008, P. R. of China

[c] The Key Laboratory of Biotechnology for Medical Plant of Jiangsu Province, Xuzhou Jiangsu 221116, P. R. of China
Fax +86(516)83403164; E-mail: xswang1974@yahoo.com

Received 6 July 2006; revised 22 August 2006

Abstract: Improved and green syntheses of 9,10-diarylacridine-1,8-dione and indenoquinoline derivatives were accomplished by the reactions of 3-anilino-5,5-dimethylcyclohex-2-enones, benzaldehydes, and 1,3-dicarbonyl compounds in the ionic liquid medium [bmim$^+$][BF$_4^-$]. Not only the substituted anilines containing electron-donating groups, but also those with electron-withdrawing groups all gave excellent yields. Furthermore, the interesting unsymmetrical 9,10-diarylacridine-1,8-dione moiety with different groups in the 3- and 6-positions and indenoquinoline derivatives were obtained and are reported here for the first time in the literature. The Knoevenagel condensation and Michael addition intermediates were obtained successfully. A possible mechanism of the reaction is discussed in detail.

Key words: 9,10-diarylacridine-1,8-diones, indenoquinolines, ionic liquids, green chemistry

Because of the toxicity and volatility of many organic solvents, room-temperature ionic liquids have been booming recently as effective solvents for 'green' processes. Especially those based on 1-alkyl-3-methylimidazolium cations[1] have shown great promise as attractive alternatives to conventional solvents, and have been the subject of considerable current interest as environmentally benign reaction media in organic synthesis, owing to their unique properties of nonvolatility, nonflammability, recyclability, and solubility in a wide range of organic components. In the past few years, a variety of ionic liquids have been demonstrated to be efficient and practical alternatives to organic solvents for many important organic transformations.[2]

1,4-Dihydropyridine and its derivatives are well-known compounds, widely prescribed as calcium β-blockers, and used for the treatment of hypertension and heart defibrillation.[3] Recently many methods have become available for the synthesis of tricyclic compounds containing 1,4-dihydropyridine moieties, such as 9-arylacridine derivatives, from aldehydes, dimedones, and ammonium acetates by traditional heating in organic solvents,[4] under triethylbenzyl ammonium chloride (TEBAC) catalysis in water,[5] under microwave irradiation,[6] or by green chemistry in ionic liquids.[7] Acridines are also important compounds possessing antitumor,[8] cytotoxic,[9] anticancer,[10] antimicrobial,[11] anti-multidrug-resistant,[12] fungicidal,[13,14] and antibacterial activity.[14]

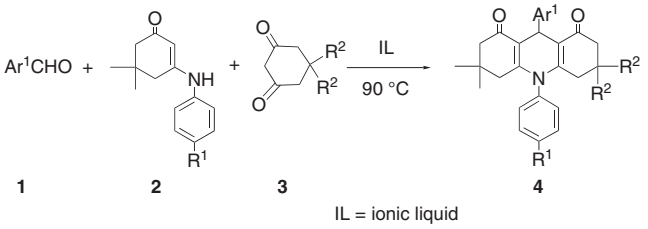
To the best of our knowledge, only a few studies on 9,10-diarylacridine-1,8-diones have been reported in the literature.[15–17] We have synthesized these compounds from benzaldehydes, dimedone, and *p*-toluidine in ethylene glycol under microwave irradiation.[15] Jin et al.[16] reported that the same reactions could be carried out catalyzed by *p*-dodecylbenzenesulfonic acid (DBSA) in water. However, it should be noted that that only *p*-toluidine was selected as substrate. The other substituted anilines either did not react, or gave low yields,[17] especially the anilines containing electron-withdrawing groups. Furthermore, the known 9,10-diarylacridine-1,8-diones are symmetrical, containing the same functional groups (methyl) on the 3- and 6-positions. To solve the abovementioned problems, and in view of the emerging importance of ionic liquids as novel reaction media, we investigated the preparation of these compounds. We here report the synthesis of symmetrical and unsymmetrical 9,10-diarylacridine-1,8-diones and indenoquinoline derivatives by a three-component, one-pot reaction of 3-anilino-5,5-dimethylcyclohex-2-enones, benzaldehydes, and 1,3-dicarbonyl compounds in an ionic liquid.

The reactions of benzaldehydes **1**, 3-anilino-5,5-dimethylcyclohex-2-enones **2**,[18] and 1,3-dicarbonyl compounds **3** in the ionic liquid medium [bmim$^+$][BF$_4^-$] (bmim = 1-butyl-3-methylimidazolium) at 90 °C gave the desired 9,10-diarylacridine-1,8-dione derivatives **4** in high yields within a few hours (Scheme 1).

We began our study of this reaction (Scheme 1) by optimizing the conditions for the preparation of 9,10-diarylacridine-1,8-dione **4a** (R^1 = R^2 = Me; Ar^1 = 4-Tol). A summary of the optimization experiment is provided in Table 1. It turned out that no reaction takes place at room temperature (Table 1, entry 1), while the reaction goes smoothly at 90 °C, giving **4a** in high yield. To find the optimum reaction time, the reaction was carried out in the ionic liquid [bmim$^+$][BF$_4^-$] for 1, 3, and 5 hours (Table 1, entries 3, 4, and 5); this gave **4a** in 86, 98, and 98% yield,

SYNTHESIS 2006, No. 24, pp 4187–4199
Advanced online publication: 13.11.2006
DOI: 10.1055/s-2006-950365; Art ID: F10406SS

respectively. Thus, a reaction temperature of 90 °C and a reaction time of three hours were chosen. Different ionic liquids, with different alkyl substituents on the methylimidazolium, and different anions, were studied, and [bmim$^+$][BF_4^-] was found to be the best medium for this reaction.



Scheme 1

Table 1 Synthesis of **4a** under Different Reaction Conditions[a]

Entry	Temp (°C)	Ionic liquid[b]	Time (h)	Yield[c] (%)
1	r.t.	[bmim$^+$][BF_4^-]	3	0
2	50	[bmim$^+$][BF_4^-]	3	56
3	90	[bmim$^+$][BF_4^-]	1	86
4	90	[bmim$^+$][BF_4^-]	3	98
5	90	[bmim$^+$][BF_4^-]	5	98
6	90	[emim$^+$]Br$^-$	3	90
7	90	[pmim$^+$]Br$^-$	3	92
8	90	[bmim$^+$]Br$^-$	3	92
9	90	[emim$^+$][BF_4^-]	3	94
10	90	[pmim$^+$][BF_4^-]	3	95

[a] Reagents and conditions for the synthesis of **4a** ($R^1 = R^2 = Me$; Ar^1 = 4-Tol): **1** (Ar^1 = 4-Tol; 1 mmol), **2** (R^1 = Me; 1 mmol), **3** (R^2 = Me; 1 mmol), ionic liquid solvent (5 mL).
[b] bmim = 1-butyl-3-methylimidazolium; emim = 1-ethyl-3-methylimidazolium; pmim = 1-methyl-3-propylimidazolium.
[c] Isolated yields.

At completion, as monitored by TLC, the reaction mixture was allowed to cool to room temperature, and the solid was isolated by filtration. The filtrate of the ionic liquid [bmim$^+$][BF_4^-] could be recycled easily by drying at 80 °C in vacuo for several hours. The recycling was investigated for the synthesis of **4a**. A summary of the effect of successive reuse of the ionic liquid is shown in Table 2. Even in the fourth run, the yield of product **4a** was fairly high.

Table 2 Study of the Reuse of the Ionic Liquid [bmim$^+$][BF_4^-][a]

Run	1	2	3	4
Yield[b] (%)	98	95	94	94

[a] Reagents and conditions for the synthesis of **4a** ($R^1 = R^2 = Me$; Ar^1 = 4-Tol): **1** (Ar^1 = 4-Tol; 1 mmol), **2** (R^1 = Me; 1 mmol), **3** (R^2 = Me; 1 mmol), ionic liquid solvent (5 mL), 90 °C, 3 h.
[b] Isolated yields.

To demonstrate the efficiency and the applicability of this method, we investigated the reactions of a variety of benzaldehydes **1**, 3-anilino-5,5-dimethylcyclohex-2-enones **2**, and 1,3-dicarbonyl compounds **3** at 90 °C in the ionic liquid medium [bmim$^+$][BF_4^-]. As shown in Table 3, a series of **1** and **2**, in which the aromatic ring contained electron-withdrawing groups (such as halo or nitro) or electron-donating groups (such as alkoxy or alkyl), reacted with **3** under the same reaction conditions to give the corresponding products **4** in high yields. We thus concluded that there were no obvious electronic or other effects of the substituents on the aromatic rings. In addition, unsymmetrical products could be prepared, e.g. 9,10-diarylacridine-1,8-diones **4** with no substituents on the 6-position (Table 3, entries 16–25).

The products **4** were completely characterized by IR and ^{1}H NMR spectroscopy and elemental analysis. The data corresponded to their structures, and the melting points of the known compounds corresponded to those reported in the literature. In addition, the X-ray diffraction analysis[19] of product **4r** was carried out to confirm its structure. The crystal structure of **4r** is shown in Figure 1.

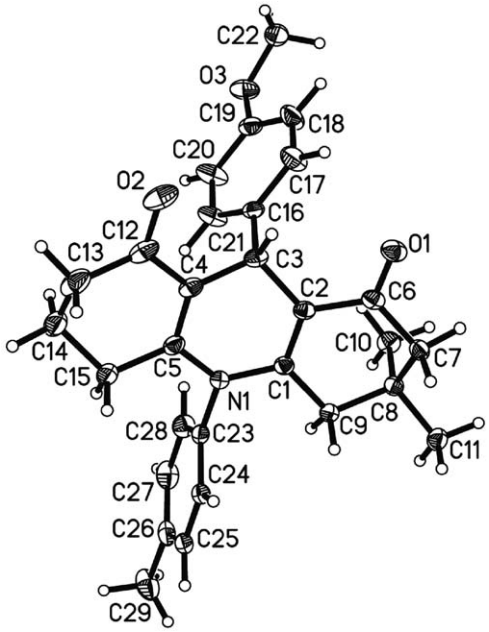


Figure 1 ORTEP drawing of **4r**

Although the detailed mechanism of the above reaction has not been clarified, the formation of 9,10-diarylacridine-1,8-dione derivatives **4** can be tentatively explained by the pathway presented in Scheme 2. Product **5** of Knoevenagel condensation may be formed as an intermediate, which cannot be obtained in this ionic liquid system. It has been reported that a 2,2′-(benzylidene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one) is obtained when a benzaldehyde is treated with 5,5-dimethylcyclohexane-1,3-dione, even when the molar ratio is kept at 1:1 in water.[20] In the following step, the Michael addition reaction may give intermediate **6**, followed by intramolecular cyclization to afford **4**.

Table 3 Synthesis of **4** in Ionic Liquid[a]

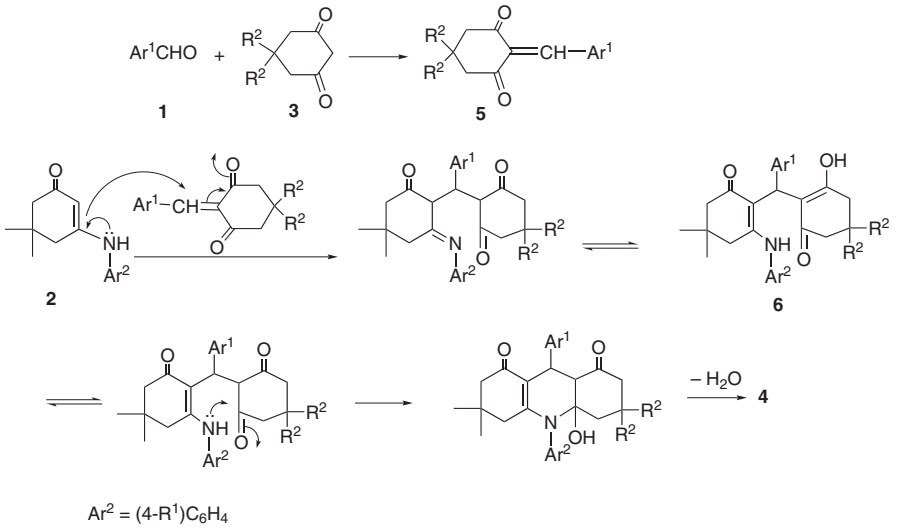
Entry	Ar^1	R^1	R^2	Time (h)	Product	Yield[b] (%)
1	4-Tol	Me	Me	3	**4a**	98
2	3,4-$(OCH_2O)C_6H_3$	Me	Me	3	**4b**	93
3	4-BrC_6H_4	Me	Me	3	**4c**	96
4	3,4-$Cl_2C_6H_3$	Me	Me	4	**4d**	95
5	4-ClC_6H_4	H	Me	3	**4e**	96
6	4-BrC_6H_4	H	Me	3	**4f**	94
7	3-$O_2NC_6H_4$	H	Me	3	**4g**	98
8	4-ClC_6H_4	F	Me	4	**4h**	95
9	4-ClC_6H_4	Cl	Me	4	**4i**	94
10	4-Tol	Br	Me	4	**4j**	95
11	4-Tol	I	Me	4	**4k**	98
12	4-ClC_6H_4	NO_2	Me	5	**4l**	94
13	2-ClC_6H_4	NO_2	Me	5	**4m**	93
14	4-BrC_6H_4	NO_2	Me	5	**4n**	97
15	4-Tol	NO_2	Me	5	**4o**	98
16	4-ClC_6H_4	Me	H	3	**4p**	94
17	4-FC_6H_4	Me	H	3	**4q**	99
18	4-Tol	Me	H	3	**4r**	98
19	4-Tol	H	H	3	**4s**	98
20	4-BrC_6H_4	H	H	3	**4t**	92
21	4-ClC_6H_4	F	H	4	**4u**	93
22	4-BrC_6H_4	Cl	H	4	**4v**	93
23	4-ClC_6H_4	Br	H	4	**4w**	94
24	3,4-$Me_2C_6H_3$	I	H	4	**4x**	97
25	4-Tol	NO_2	H	5	**4y**	98

[a] Reagents and conditions: **1** (1 mmol), **2** (1 mmol), **3** (1 mmol), ionic liquid (5 mL).
[b] Isolated yields.

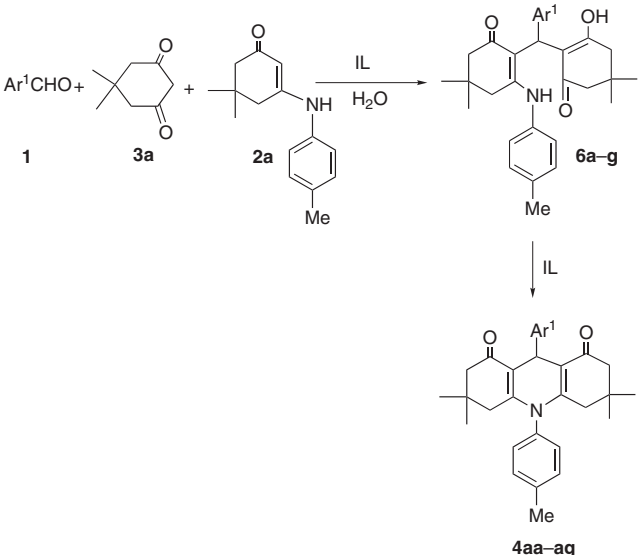
To prove the mechanism, we decided to perform the reaction in the ionic liquid in the presence of water, as we think intermediates **6** may be obtained in that case. As expected, aniline derivative **6a** (Ar^1 = Ph) was obtained, in good yield, from the reaction between 5,5-dimethyl-3-(4-methylanilino)cyclohex-2-enone (**2a**), benzaldehyde (**1**, Ar^1 = Ph), and 5,5-dimethylcyclohexane-1,3-dione (**3a**) in a medium of ionic liquid and water at 90 °C (Scheme 3, Table 4). The structure of compound **6a** was also confirmed by X-ray crystal structure analysis (Figure 2).[21] We were pleased to find that **6a** smoothly gave the corresponding 9,10-diarylacridine-1,8-dione **4aa** by intramolecular cyclization as expected (Table 4, entry 1). This result possibly indicates that a Michael addition reaction took place in the above reaction.

Encouraged by this result, we synthesized a number of derivatives **6b–g** by this method (Scheme 3, Table 4). Similarly, compounds **6b–g** smoothly reacted in ionic liquid to give the corresponding 9,10-diarylacridine-1,8-diones **4ab–ag** in high yields (Table 4, entries 2–7).

We were interested in the intermediate 2-benzylidenecyclohexane-1,3-dione **5**, and therefore decided to prepare 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione, a cyclopentanedione equivalent of **5**, from indene-1,3-dione (**7**) as the 1,3-dicarbonyl compound. The Knoevenagel condensation product, 2-(4-bromobenzylidene)-1*H*-in-



Scheme 2



Scheme 3

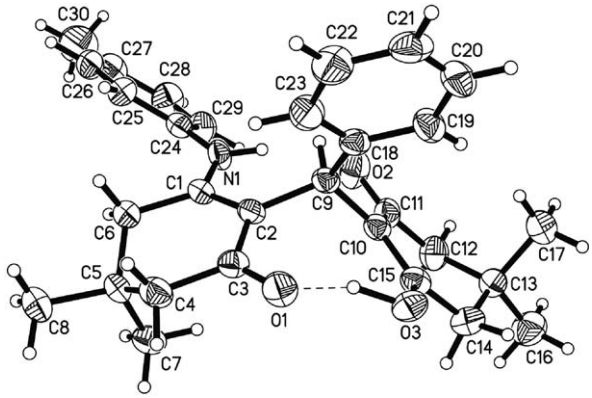


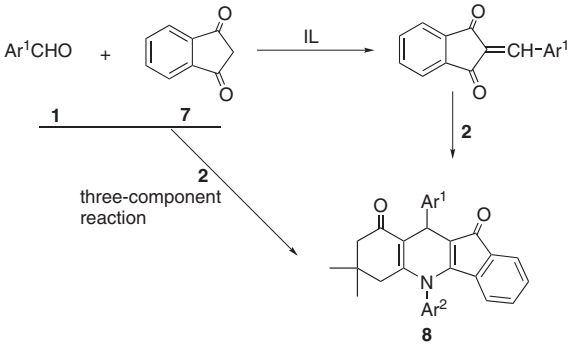
Figure 2 ORTEP drawing of **6a**

dene-1,3(2*H*)-dione was obtained in 98% yield when 4-bromobenzaldehyde (**1**, $Ar^1 = 4\text{-}BrC_6H_4$) was treated with indene-1,3-dione in ionic liquid (Scheme 4). As expected, intermediate smoothly reacted with **2a** to give the corresponding indeno[1,2-*b*]quinolinedione **8a** (Scheme 4,

Table 4 Synthesis of **6a–f** and **4aa–af**

Ar^1	Product **6**	Time (h)	Yield[a] (%)	Product **4**	Time (h)	Yield[a] (%)
Ph	**6a**	15	83	**4aa**	2	97
$2\text{-}ClC_6H_4$	**6b**	12	89	**4ab**	1	96
$3\text{-}ClC_6H_4$	**6c**	12	88	**4ac**	1	95
$4\text{-}ClC_6H_4$	**6d**	14	85	**4ad**	1	98
4-Tol	**6e**	16	82	**4ae**	3	96
$3\text{-}O_2NC_6H_4$	**6f**	12	86	**4af**	1	98
$4\text{-}FC_6H_4$	**6g**	10	88	**4ag**	1	98

[a] Isolated yields.



Scheme 4

Table 5). This result suggests that a Knoevenagel condensation took place during the reaction. On the basis of this discovery, a series of indenoquinolines **8a–i** was synthesized by one-pot three-component reactions between aromatic aldehydes **1**, 3-anilino-5,5-dimethylcyclohex-2-enones **2**, and indene-1,3-dione (**7**) in the ionic liquid medium [bmim$^+$][BF_4^-]. The results are summarized in Table 5. The structure of product **8b** was confirmed by X-ray diffraction analysis,[22] and is shown in Figure 3.

Table 5 Synthesis of **8a–8i**

Product **8**	Ar^1	Ar^2	Time (h)	Yield (%)
8a	4-BrC_6H_4	4-Tol	5	98
8b	4-FC_6H_4	4-Tol	5	96
8c	4-BrC_6H_4	4-BrC_6H_4	6	97
8d	4-FC_6H_4	4-BrC_6H_4	6	98
8e	2,4-$Cl_2C_6H_3$	4-BrC_6H_4	6	95
8f	3-ClC_6H_4	1-naphthyl	7	96
8g	2-ClC_6H_4	1-naphthyl	7	96
8h	4-Tol	1-naphthyl	8	95
8i	4-FC_6H_4	1-naphthyl	8	97

In conclusion, we have developed a novel synthetic method for the synthesis of 9,10-diarylacridine and indenoquinoline derivatives in excellent yields by using the ionic liquid [bmim$^+$][BF_4^-] as medium. Excellent yields were obtained, not only for anilines substituted with electron-donating groups, but also for ones containing electron-withdrawing groups, in contrast to other methods.[15–17] Furthermore, the interesting unsymmetrical 9,10-diarylacridine-1,8-dione moiety with different groups in 3- and 6-positions were obtained. The ionic liquid medium [bmim$^+$][BF_4^-] was chosen as a green solvent, which could be reused for several rounds without significant loss of activity.

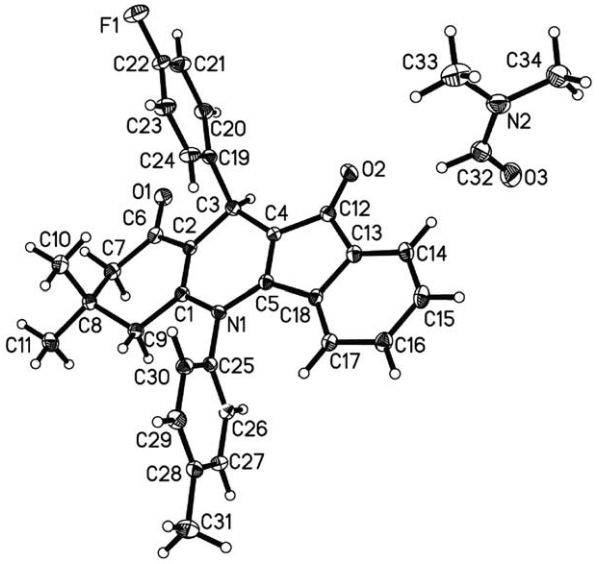


Figure 3 ORTEP drawing of **8b** with DMF solvate

Melting points were determined in open capillaries and are uncorrected. IR spectra of samples prepared in KBr pellets were recorded on a TENSOR 27 spectrometer. ^{1}H NMR spectra of DMSO-d_6 or $CDCl_3$ solutions with TMS as internal standard were obtained on a Bruker-400 spectrometer. Elemental analyses were carried out on a Carlo Erba 1110 analyzer. X-ray crystal-structure determinations were determined on a Rigaku Mercury or CCD area diffractometer.

9,10-Diarylacridine-1,8-diones 4a–y; General Procedure

A mixture of benzaldehyde **1** (1.0 mmol), 3-anilino-5,5-dimethylcyclohex-2-enone **2** (1.0 mmol), 1,3-dicarbonyl compound **3** (1.0 mmol), and ionic liquid medium [bmim$^+$][BF_4^-] (5 mL) was stirred at 90 °C for several hours to complete the reaction (monitored by TLC). The mixture was cooled to r.t. and the yellow solid was collected by filtration. The filtrate of the ionic liquid [bmim$^+$][BF_4^-] could then be recycled after drying at 80 °C for several hours in vacuo. The crude product was washed with H_2O and purified by recrystallization from DMF–H_2O; this gave **4**.

3,3,6,6-Tetramethyl-9,10-bis(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4a)

Pale yellow crystals; mp 296–297 °C (Lit.[15] 296–297 °C).

9-(1,3-Benzodioxol-5-yl)-3,3,6,6-tetramethyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4b)

Pale yellow crystals; mp 278–280 °C (Lit.[15] 278–279 °C).

9-(4-Bromophenyl)-3,3,6,6-tetramethyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4c)

Pale yellow crystals; mp 270–272 °C.

IR (KBr): 3035, 2956, 2869, 1638, 1576, 1511, 1483, 1450, 1405, 1361, 1301, 1278, 1221, 1176, 1144, 1211, 1069, 1009, 841 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.71 (s, 6 H, 2 CH_3), 0.87 (s, 6 H, 2 CH_3), 1.77 (d, J = 17.2 Hz, 2 H, 2 CH), 2.00 (d, J = 16.0 Hz, 2 H, 2 CH), 2.17–2.22 (m, 4 H, 4 CH), 2.42 (s, 3 H, CH_3), 5.00 (s, 1 H, CH), 7.25–7.31 (m, 4 H, ArH), 7.41 (d, J = 8.8 Hz, 2 H, ArH), 7.44 (d, J = 8.4 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 21.0, 26.3, 29.4, 32.0, 32.1, 41.1, 49.6, 112.6, 118.9, 130.0, 131.0, 135.9, 139.1, 145.8, 151.0, 195.3.

Anal. Calcd for $C_{30}H_{32}BrNO_2$: C, 69.50; H, 6.22; N, 2.70. Found: C, 69.38; H, 6.30; N, 2.78.

9-(3,4-Dichlorophenyl)-3,3,6,6-tetramethyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4d)
Pale yellow crystals; mp 251–253 °C.

IR (KBr): 3035, 2957, 2869, 1642, 1573, 1510, 1469, 1359, 1301, 1279, 1218, 1142, 1121, 1023, 1000, 879, 834, 735 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.72 (s, 6 H, 2 CH_3), 0.88 (s, 6 H, 2 CH_3), 1.79 (d, *J* = 17.6 Hz, 2 H, 2 CH), 2.02 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.20 (d, *J* = 16.8 Hz, 4 H, 4 CH), 2.42 (s, 3 H, CH_3), 4.99 (s, 1 H, CH), 7.22–7.33 (m, 4 H, ArH), 7.41–7.45 (m, 2 H, ArH), 7.54 (d, *J* = 8.4 Hz, 1 H, ArH).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 21.3, 26.6, 29.6, 32.46, 32.48, 41.4, 49.8, 112.4, 128.2, 128.7, 130.2, 130.7, 130.9, 136.0, 139.5, 147.7, 151.6, 195.6.

Anal. Calcd for $C_{30}H_{31}Cl_2NO_2$: C, 70.86; H, 6.15; N, 2.75. Found: C, 70.70; H, 6.25; N, 2.66.

9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-10-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4e)
Pale yellow crystals; mp 244–246 °C.

IR (KBr): 3063, 2956, 2870, 1634, 1575, 1489, 1452, 1409, 1361, 1299, 1274, 1224, 1141, 1122, 1088, 1014, 1000, 942, 920, 839, 770, 705 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.71 (s, 6 H, 2 CH_3), 0.88 (s, 6 H, 2 CH_3), 1.75 (d, *J* = 17.6 Hz, 2 H, 2 CH), 2.01 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.20 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.21 (d, *J* = 17.6 Hz, 2 H, 2 CH), 5.03 (s, 1 H, CH), 7.29–7.35 (m, 4 H, ArH), 7.43 (b, 2 H, ArH), 7.58–7.64 (m, 3 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.3, 29.4, 31.9, 32.1, 41.1, 49.6, 112.7, 128.1, 129.57, 129.64, 130.2, 130.4, 138.5, 145.4, 150.7, 195.3.

Anal. Calcd for $C_{29}H_{30}ClNO_2$: C, 75.72; H, 6.57; N, 3.04. Found: C, 75.58; H, 6.70; N, 3.12.

9-(4-Bromophenyl)-3,3,6,6-tetramethyl-10-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4f)
Pale yellow crystals; mp 238–240 °C.

IR (KBr): 3061, 2959, 2869, 1659, 1638, 1576, 1485, 1467, 1451, 1406, 1360, 1300, 1277, 1225, 1145, 1122, 1093, 1067, 1008, 921, 840, 771, 706, 659 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.71 (s, 6 H, 2 CH_3), 0.88 (s, 6 H, 2 CH_3), 1.75 (d, *J* = 17.6 Hz, 2 H, 2 CH), 2.01 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.18–2.23 (m, 4 H, 4 CH), 5.01 (s, 1 H, CH), 7.27 (d, *J* = 8.4 Hz, 2 H, ArH), 7.44–7.46 (m, 4 H, ArH), 7.58–7.64 (m, 3 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.3, 29.4, 32.0, 32.1, 41.1, 49.6, 112.6, 118.9, 129.6, 130.0, 130.2, 131.0, 138.5, 145.8, 150.7, 195.3.

Anal. Calcd for $C_{29}H_{30}BrNO_2$: C, 69.05; H, 5.99; N, 2.78. Found: C, 68.91; H, 5.96; N, 2.90.

3,3,6,6-Tetramethyl-9-(3-nitrophenyl)-10-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4g)
Pale yellow crystals; mp 245–247 °C.

IR (KBr): 3062, 2951, 2871, 1638, 1574, 1526, 1490, 1472, 1452, 1422, 1361, 1301, 1262, 1222, 1123, 1091, 1020, 1002, 922, 899, 818, 733, 708, 688 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.70 (s, 6 H, 2 CH_3), 0.87 (s, 6 H, 2 CH_3), 1.79 (d, *J* = 17.6 Hz, 2 H, 2 CH), 2.02 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.23 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.25 (d, *J* = 17.6 Hz, 2 H, 2 CH), 5.15 (s, 1 H, CH), 7.45–7.67 (m, 4 H, ArH), 7.79 (d, *J* = 8.0 Hz, 1 H, ArH), 8.02 (dd, *J* = 8.0, 1.2 Hz, 1 H, ArH), 8.15 (s, 1 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.2, 29.3, 32.2, 32.6, 41.1, 49.5, 112.3, 121.2, 122.2, 129.8, 129.9, 130.2, 134.4, 138.3, 147.6, 148.4, 151.3, 195.4.

Anal. Calcd for $C_{29}H_{30}N_2O_4$: C, 74.02; H, 6.43; N, 5.95. Found: C, 74.10; H, 6.44; N, 5.82.

9-(4-Chlorophenyl)-10-(4-fluorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4h)
Pale yellow crystals; mp >300 °C.

IR (KBr): 3061, 2956, 2933, 2869, 1639, 1578, 1507, 1486, 1412, 1361, 1302, 1279, 1262, 1177, 1144, 1119, 1089, 1013, 980, 934, 888, 840, 805, 780, 743, 714 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.72 (s, 6 H, 2 CH_3), 0.89 (s, 6 H, 2 CH_3), 1.77 (d, *J* = 17.2 Hz, 2 H, 2 CH), 2.01 (d, *J* = 16.4 Hz, 2 H, 2 CH), 2.17–2.22 (m, 4 H, 4 CH), 5.01 (s, 1 H, CH), 7.29–7.53 (m, 8 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.3, 29.4, 31.9, 32.1, 41.1, 49.6, 112.8, 117.0, 128.1, 129.6, 130.4, 134.8, 145.3, 150.8, 160.8, 163.3, 195.3.

Anal. Calcd for $C_{29}H_{29}ClFNO_2$: C, 72.87; H, 6.12; N, 2.93. Found: C, 72.80; H, 6.22; N, 2.88.

9,10-Bis(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4i)
Pale yellow crystals; mp 284–286 °C.

IR (KBr): 3051, 2958, 2869, 1639, 1578, 1490, 1406, 1361, 1302, 1278, 1263, 1177, 1145, 1121, 1089, 1014, 888, 839, 739, 713 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.72 (s, 6 H, 2 CH_3), 0.89 (s, 6 H, 2 CH_3), 1.78 (d, *J* = 17.6 Hz, 2 H, 2 CH), 2.01 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.19 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.20 (d, *J* = 17.6 Hz, 2 H, 2 CH), 5.01 (s, 1 H, CH), 7.30–7.49 (m, 6 H, ArH), 7.68 (d, *J* = 8.8 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.2, 29.4, 31.9, 32.2, 41.1, 49.7, 112.8, 128.1, 129.6, 130.5, 134.2, 137.4, 145.2, 150.5, 195.3.

Anal. Calcd for $C_{29}H_{29}Cl_2NO_2$: C, 70.44; H, 5.91; N, 2.83. Found: C, 70.28; H, 6.05; N, 2.90.

10-(4-Bromophenyl)-9-(4-methoxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4j)
Pale yellow crystals; mp 247–248 °C.

IR (KBr): 3050, 3006, 2956, 2836, 1641, 1608, 1580, 1508, 1489, 1472, 1440, 1361, 1299, 1260, 1176, 1141, 1121, 1104, 1037, 1013, 999, 920, 886, 863, 834, 738 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.73 (s, 6 H, 2 CH_3), 0.89 (s, 6 H, 2 CH_3), 1.76 (d, *J* = 17.6 Hz, 2 H, 2 CH), 2.00 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.18 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.19 (d, *J* = 17.6 Hz, 2 H, 2 CH), 3.69 (s, 3 H, CH_3O), 4.97 (s, 1 H, CH), 6.80 (d, *J* = 8.4 Hz, 2 H, ArH), 7.20 (d, *J* = 8.4 Hz, 2 H, ArH), 7.39 (b, 2 H, ArH), 7.81 (d, *J* = 8.4 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.2, 29.4, 31.1, 32.1, 41.1, 49.8, 55.0, 113.4, 113.5, 122.7, 128.6, 133.2, 138.0, 138.6, 150.0, 157.5, 195.3.

Anal. Calcd for $C_{30}H_{32}BrNO_3$: C, 67.41; H, 6.03; N, 2.62. Found: C, 67.52; H, 6.22; N, 2.66.

10-(4-Iodophenyl)-9-(4-methoxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4k)
Pale yellow crystals; mp 259–262 °C.

IR (KBr): 3031, 2956, 2871, 1644, 1581, 1508, 1442, 1422, 1368, 1301, 1261, 1218, 1140, 1102, 1033, 1009, 850, 833 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.73 (s, 6 H, 2 CH_3), 0.89 (s, 6 H, 2 CH_3), 1.76 (d, *J* = 17.2 Hz, 2 H, 2 CH), 2.00 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.18 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.19 (d, *J* = 17.6 Hz, 2 H, 2 CH), 3.69 (s, 3 H, CH_3O), 4.97 (s, 1 H, CH), 6.80 (d, *J* = 8.4 Hz, 2 H, ArH), 7.20 (d, *J* = 8.4 Hz, 4 H, ArH), 7.97 (d, *J* = 8.4 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.2, 29.4, 31.2, 41.1, 49.8, 55.0, 96.2, 113.4, 113.5, 128.6, 138.4, 138.6, 139.1, 150.0, 157.5, 195.3.

Anal. Calcd for $C_{30}H_{32}INO_3$: C, 61.97; H, 5.55; N, 2.41. Found: C, 61.82; H, 5.70; N, 2.54.

9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-10-(4-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4l)
Pale yellow crystals; mp >300 °C.

IR (KBr): 3067, 2958, 2871, 1640, 1608, 1592, 1525, 1487, 1362, 1346, 1298, 1278, 1261, 1224, 1178, 1087, 1013, 921, 863, 842, 708 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.72 (s, 6 H, 2 CH_3), 0.89 (s, 6 H, 2 CH_3), 1.79 (d, *J* = 17.2 Hz, 2 H, 2 CH), 2.03 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.21 (d, *J* = 16.8 Hz, 4 H, 4 CH), 5.02 (s, 1 H, CH), 7.31 (d, *J* = 8.4 Hz, 2 H, ArH), 7.34 (d, *J* = 8.4 Hz, 2 H, ArH), 7.77 (d, *J* = 8.8 Hz, 2 H, ArH), 8.45 (d, *J* = 8.8 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.2, 29.3, 31.9, 32.3, 41.0, 49.7, 113.1, 125.4, 128.0, 128.1, 129.7, 131.7, 144.2, 145.1, 150.0, 195.4.

Anal. Calcd for $C_{29}H_{29}ClN_2O_4$: C, 68.97; H, 5.79; N, 5.55. Found: C, 68.79; H, 5.92; N, 5.50.

9-(2-Chlorophenyl)-3,3,6,6-tetramethyl-10-(4-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4m)
Pale yellow crystals; mp >300 °C.

IR (KBr): 3065, 2960, 2871, 1643, 1592, 1579, 1524, 1425, 1366, 1345, 1303, 1263, 1224, 1177, 1145, 1014, 862, 741, 707 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.74 (s, 6 H, 2 CH_3), 0.87 (s, 6 H, 2 CH_3), 1.76 (d, *J* = 17.6 Hz, 2 H, 2 CH), 1.96 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.12 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.16 (d, *J* = 17.2 Hz, 2 H, 2 CH), 5.27 (s, 1 H, CH), 7.10–7.14 (m, 1 H, ArH), 7.23–7.28 (m, 2 H, ArH), 7.52 (d, *J* = 7.6 Hz, 1 H, ArH), 7.76 (d, *J* = 8.8 Hz, 2 H, ArH), 8.45 (d, *J* = 8.8 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 26.7, 29.6, 32.3, 34.6, 42.1, 49.9, 112.8, 125.4, 126.2, 127.5, 130.1, 131.3, 133.3, 133.8, 141.6, 144.8, 147.9, 149.3, 195.6.

Anal. Calcd for $C_{29}H_{29}ClN_2O_4$: C, 68.97; H, 5.79; N, 5.55. Found: C, 68.82; H, 5.90; N, 5.59.

9-(4-Bromophenyl)-3,3,6,6-tetramethyl-10-(4-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4n)
Pale yellow crystals; mp 291–293 °C.

IR (KBr): 3049, 2958, 2969, 1642, 1579, 1487, 1361, 1301, 1263, 1177, 1144, 1122, 1100, 1069, 1010, 946, 921, 888, 838, 773, 736, 711, 669, 615 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.73 (s, 6 H, 2 CH_3), 0.89 (s, 6 H, 2 CH_3), 1.78 (d, *J* = 17.6 Hz, 2 H, 2 CH), 2.01 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.21 (d, *J* = 17.6 Hz, 2 H, 2 CH), 2.27 (d, *J* = 16.0 Hz, 2 H, 2 CH), 5.01 (s, 1 H, CH), 7.13 (d, *J* = 8.4 Hz, 2 H, ArH), 7.42 (d, *J* = 8.4 Hz, 2 H, ArH), 7.77 (d, *J* = 8.8 Hz, 2 H, ArH), 8.45 (d, *J* = 8.8 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.5, 28.2, 31.8, 32.6, 41.3, 47.1, 113.3, 114.6, 119.3, 125.7, 129.3, 130.0, 131.1, 131.3, 144.5, 148.2, 195.7.

Anal. Calcd for $C_{29}H_{29}BrN_2O_4$: C, 63.39; H, 5.32; N, 5.10. Found: C, 63.21; H, 5.60; N, 5.12.

9-(4-Methoxyphenyl)-3,3,6,6-tetramethyl-10-(4-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4o)
Pale yellow crystals; mp >300 °C.

IR (KBr): 3070, 2959, 2871, 2836, 1667, 1608, 1591, 1532, 1508, 1491, 1465, 1390, 1361, 1301, 1262, 1170, 1143, 1032, 1008, 889, 865, 850, 833, 706, 656 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.73 (s, 6 H, 2 CH_3), 0.88 (s, 6 H, 2 CH_3), 1.77 (d, *J* = 17.6 Hz, 2 H, 2 CH), 2.02 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.19 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.20 (d, *J* = 17.6 Hz, 2 H, 2 CH), 3.70 (s, 3 H, CH_3O), 4.98 (s, 1 H, CH), 8.01 (d, *J* = 8.4 Hz, 2 H, ArH), 7.23 (d, *J* = 8.4 Hz, 2 H, ArH), 7.24 (d, *J* = 8.4 Hz, 2 H, ArH), 8.44 (d, *J* = 8.4 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 26.9, 29.7, 31.8, 32.6, 42.0, 50.1, 55.2, 113.6, 115.5, 125.4, 128.8, 131.2, 138.1, 145.0, 147.97, 148.03, 157.9, 195.7.

Anal. Calcd for $C_{30}H_{32}N_2O_5$: C, 71.98; H, 6.44; N, 5.60. Found: C, 71.82; H, 6.58; N, 5.52.

9-(4-Chlorophenyl)-3,3-dimethyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4p)
Pale yellow crystals; mp 244–245 °C.

IR (KBr): 3054, 2957, 2868, 1644, 1573, 1510, 1485, 1448, 1427, 1408, 1361, 1292, 1266, 1183, 1149, 1134, 1093, 1014, 977, 915, 857, 836, 769, 723 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.71 (s, 3 H, CH_3), 0.88 (s, 3 H, CH_3), 1.61–2.27 (m, 10 H, 5 CH_2), 2.41 (s, 3 H, CH_3), 5.06 (s, 1 H, CH), 7.24–7.41 (m, 8 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.85, 20.91, 26.2, 27.9, 29.5, 31.5, 32.1, 36.3, 41.1, 49.7, 112.7, 113.5, 128.1, 129.5, 130.0, 130.4, 130.8, 135.9, 139.1, 145.6, 150.8, 153.1, 195.4, 195.6.

Anal. Calcd for $C_{28}H_{28}ClNO_2$: C, 75.41; H, 6.33; N, 3.14. Found: C, 75.29; H, 6.55; N, 3.12.

9-(4-Fluorophenyl)-3,3-dimethyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4q)
Pale yellow crystals; mp 261–264 °C.

IR (KBr): 3062, 2960, 2870, 1644, 1571, 1503, 1449, 1429, 1415, 1361, 1292, 1265, 1183, 1151, 1133, 1089, 1041, 1017, 977, 916, 894, 848, 809, 769, 736, 723, 659 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.71 (s, 3 H, CH_3), 0.88 (s, 3 H, CH_3), 1.50–2.23 (m, 10 H, 5 CH_2), 2.41 (s, 3 H, CH_3), 5.07 (s, 1 H, CH), 7.06 (t, *J* = 8.8 Hz, 2 H, ArH), 7.23–7.41 (m, 6 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.86, 20.91, 26.2, 27.9, 29.5, 31.2, 32.1, 36.4, 41.1, 49.7, 113.0, 113.8, 114.7, 114.9, 129.3, 129.4, 136.0, 139.1, 142.88, 142.91, 150.7, 153.0, 159.4, 161.8, 195.4, 195.6.

Anal. Calcd for $C_{28}H_{28}FNO_2$: C, 78.30; H, 6.57; N, 3.26. Found: C, 78.29; H, 6.68; N, 3.20.

9-(4-Methoxyphenyl)-3,3-dimethyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4r)
Pale yellow crystals; mp 254–256 °C.

IR (KBr): 3035, 2943, 2870, 2840, 1634, 1573, 1509, 1455, 1423, 1361, 1302, 1289, 1264, 1225, 1187, 1149, 1123, 1032, 979, 894, 831, 767, 737, 658 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.72 (s, 3 H, CH_3), 0.88 (s, 3 H, CH_3), 1.60–2.22 (m, 10 H, 5 CH_2), 2.41 (s, 3 H, CH_3), 3.69 (s, 3 H, CH_3O), 5.02 (s, 1 H, CH), 6.80 (d, *J* = 8.8 Hz, 2 H, ArH), 7.20 (d, *J* = 8.4 Hz, 3 H, ArH), 7.39 (d, *J* = 8.0 Hz, 3 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.9, 26.2, 27.8, 29.5, 30.7, 32.1, 36.4, 41.1, 49.8, 50.1, 113.4, 113.5, 114.3, 128.5, 128.9, 130.8, 136.1, 139.0, 150.3, 152.6, 157.5, 195.4, 195.6.

Anal. Calcd for $C_{29}H_{31}NO_3$: C, 78.88; H, 7.08; N, 3.17. Found: C, 78.76; H, 7.29; N, 3.25.

9-(4-Methoxyphenyl)-3,3-dimethyl-10-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4s)
Pale yellow crystals; mp 256–258 °C.

IR (KBr): 3063, 2978, 2953, 2870, 1634, 1570, 1507, 1493, 1454, 1361, 1286, 1226, 1148, 1136, 1103, 1031, 980, 892, 834, 707 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.73 (s, 3 H, CH_3), 0.88 (s, 3 H, CH_3), 1.60–1.62 (m, 1 H, CH), 1.72–2.03 (m, 4 H, 4 CH), 2.16–2.26 (m, 5 H, 5 CH), 3.70 (s, 3 H, CH_3O), 5.03 (s, 1 H, CH), 6.81 (d, *J* = 8.4 Hz, 2 H, ArH), 7.21 (d, *J* = 8.4 Hz, 2 H, ArH), 7.40–7.62 (m, 5 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.9, 26.3, 27.9, 30.8, 32.1, 36.4, 41.1, 49.8, 55.0, 113.4, 113.5, 114.3, 128.6, 129.5, 130.3, 138.7, 139.0, 150.1, 152.3, 157.5, 195.4, 195.6.

Anal. Calcd for $C_{28}H_{29}NO_3$: C, 78.66; H, 6.84; N, 3.28. Found: C, 78.53; H, 6.85; N, 3.26.

9-(4-Bromophenyl)-3,3-dimethyl-10-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4t)
Pale yellow crystals; mp 239–240 °C.

IR (KBr): 3048, 2959, 2928, 2884, 1636, 1592, 1574, 1490, 1452, 1361, 1285, 1267, 1185, 1136, 1068, 980, 834, 707 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.71 (s, 3 H, CH_3), 0.87 (s, 3 H, CH_3), 1.60–1.64 (m, 1 H, CH), 1.73–2.04 (m, 4 H, 4 CH), 2.18–2.28 (m, 5 H, 5 CH), 5.06 (s, 1 H, CH), 7.26 (d, *J* = 7.6 Hz, 2 H, ArH), 7.43–7.60 (m, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.9, 26.2, 27.9, 29.4, 31.6, 32.6, 36.3, 41.1, 49.7, 112.7, 113.5, 119.0, 129.6, 130.0, 130.2, 131.0, 138.5, 146.0, 150.7, 152.9, 195.4, 195.6.

Anal. Calcd for $C_{27}H_{26}BrNO_2$: C, 68.07; H, 5.50; N, 2.94. Found: C, 67.90; H, 5.65; N, 2.90.

9-(4-Chlorophenyl)-10-(4-fluorophenyl)-3,3-dimethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4u)
Pale yellow crystals; mp 237–239°C.

IR (KBr): 3064, 2957, 2870, 1683, 1632, 1572, 1506, 1454, 1411, 1362, 1308, 1286, 1265, 1186, 1138, 1088, 1013, 981, 936, 894, 855, 832, 792, 771 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.73 (s, 3 H, CH_3), 0.89 (s, 3 H, CH_3), 1.62–1.65 (m, 1 H, CH), 1.75–1.86 (m, 2 H, 2 CH), 1.94–2.04 (m, 2 H, 2 CH), 2.17–2.29 (m, 5 H, 5 CH), 5.07 (s, 1 H, CH), 7.28–7.33 (m, 4 H, ArH), 7.43 (b, 3 H, ArH), 7.57 (b, 1 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 21.1, 26.5, 28.2, 29.7, 31.8, 32.4, 36.6, 41.4, 50.0, 113.2, 113.9, 128.4, 129.9, 130.8, 135.06, 135.09, 145.8, 151.0, 153.3, 161.1, 162.8, 195.7, 195.9.

Anal. Calcd for $C_{27}H_{25}ClFNO_2$: C, 72.07; H, 5.60; N, 3.11. Found: C, 72.00; H, 5.67; N, 3.15.

9-(4-Bromophenyl)-10-(4-chlorophenyl)-3,3-dimethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4v)
Pale yellow crystals; mp 249–251 °C.

IR (KBr): 3052, 2957, 2868, 1640, 1576, 1489, 1404, 1361, 1285, 1268, 1227, 1185, 1138, 1088, 1069, 1010, 981, 937, 858, 828, 746 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.73 (s, 3 H, CH_3), 0.89 (s, 3 H, CH_3), 1.61–1.66 (m, 1 H, CH), 1.76–1.86 (m, 2 H, 2 CH), 1.92–2.04 (m, 2 H, 2 CH), 2.16–2.22 (m, 5 H, 5 CH), 5.05 (s, 1 H, CH), 7.26 (d, *J* = 8.4 Hz, 2 H, ArH), 7.42 (d, *J* = 8.4 Hz, 2 H, ArH), 7.50 (b, 2 H, ArH), 7.67 (d, *J* = 8.4 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.9, 26.2, 27.9, 29.4, 31.6, 32.1, 36.3, 41.0, 49.7, 106.2, 112.8, 113.6, 119.0, 130.0, 131.0, 134.1, 137.4, 145.9, 150.4, 152.7, 195.4, 195.6.

Anal. Calcd for $C_{27}H_{25}BrClNO_2$: C, 63.48; H, 4.93; N, 2.74. Found: C, 63.34; H, 5.12; N, 2.78.

10-(4-Bromophenyl)-9-(4-chlorophenyl)-3,3-dimethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4w)
Pale yellow crystals; mp 257–259 °C.

IR (KBr): 3059, 2958, 2867, 1650, 1579, 1486, 1448, 1405, 1293, 1269, 1184, 1135, 1092, 1066, 1011, 978, 916, 851, 831, 769, 726, 626, 579 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.72 (s, 3 H, CH_3), 0.89 (s, 3 H, CH_3), 1.62–1.64 (m, 1 H, CH), 1.73–1.83 (m, 2 H, 2 CH), 1.90–2.04 (m, 2 H, 2 CH), 2.17–2.22 (m, 5 H, 5 CH), 5.06 (s, 1 H, CH), 7.29 (d, *J* = 8.4 Hz, 2 H, ArH), 7.31 (d, *J* = 8.4 Hz, 2 H, ArH), 7.44 (b, 2 H, ArH), 7.80 (d, *J* = 8.8 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.9, 26.2, 27.9, 31.5, 32.1, 36.3, 41.1, 49.7, 112.9, 113.7, 122.8, 129.6, 130.5, 133.1, 137.8, 145.4, 150.3, 152.7, 195.4, 195.6.

Anal. Calcd for $C_{27}H_{25}BrClNO_2$: C, 63.48; H, 4.93; N, 2.74. Found: C, 63.38; H, 5.10; N, 2.98.

9-(3,4-Dimethylphenyl)-10-(4-iodophenyl)-3,3-dimethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4x)
Pale yellow crystals; mp 258–261 °C.

IR (KBr): 3014, 2957, 2884, 1650, 1574, 1483, 1454, 1418, 1361, 1284, 1266, 1186, 1136, 1121, 1008, 978, 936, 843, 793, 736 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.75 (s, 3 H, CH_3), 0.90 (s, 3 H, CH_3), 1.57–1.62 (m, 1 H, CH), 1.77–1.84 (m, 2 H, 2 CH), 1.90–2.03 (m, 2 H, 2 CH), 2.18–2.25 (m, 10 H, 2 CH_3 + 4 CH), 5.00 (s, 1 H, CH), 6.98–7.04 (m, 3 H, ArH), 7.21 (b, 2 H, ArH), 7.96 (d, *J* = 8.4 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 19.1, 19.8, 20.9, 26.2, 27.9, 29.5, 31.1, 32.2, 36.4, 41.0, 49.8, 96.1, 113.4, 114.5, 124.8, 129.1, 129.4, 132.3, 133.6, 135.3, 138.5, 139.0, 144.1, 150.0, 152.0, 195.4, 195.6.

Anal. Calcd for $C_{29}H_{30}INO_2$: C, 63.16; H, 5.48; N, 2.54. Found: C, 63.10; H, 5.63; N, 2.65.

9-(4-Methoxyphenyl)-3,3-dimethyl-10-(4-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4y)
Pale yellow crystals; mp 259–261 °C.

IR (KBr): 3065, 2954, 1645, 1592, 2527, 1507, 1455, 1362, 1268, 1228, 1137, 1037, 980, 939, 863, 831 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.74 (s, 3 H, CH_3), 0.89 (s, 3 H, CH_3), 1.61–1.66 (m, 1 H, CH), 1.75–1.85 (m, 2 H, 2 CH), 1.92–2.05 (m, 2 H, 2 CH), 2.17–2.28 (m, 5 H, 5 CH), 3.70 (s, 3 H, CH_3O), 5.03 (s, 1 H, CH), 6.81 (d, *J* = 8.4 Hz, 2 H, ArH), 7.22 (d, *J* = 8.4 Hz, 2 H, ArH), 7.75 (d, *J* = 8.8 Hz, 2 H, ArH), 8.43 (d, *J* = 8.8 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.9, 26.2, 27.9, 29.4, 30.8, 32.2, 36.4, 41.0, 49.8, 55.1, 113.5, 113.8, 114.7, 125.3, 128.7, 131.7, 138.7, 144.4, 147.8, 149.4, 151.6, 157.5, 195.4, 195.6.

Anal. Calcd for $C_{28}H_{28}N_2O_5$: C, 71.17; H, 5.97; N, 5.93. Found: C, 71.02; H, 6.12; N, 6.05.

Aniline Derivatives 6a–g; General Procedure

A mixture of benzaldehyde **1** (3.0 mmol), 5,5-dimethyl-3-(4-methylanilino)cyclohex-2-enone (**2a**, 3.0 mmol), and 5,5-dimethylcyclohexane-1,3-dione (**3a**, 3.0 mmol) in a mixture of the ionic liquid [bmim$^+$][BF_4^-] (5.0 mL) and H_2O (5.0 mL) was stirred at 90 °C for several hours to complete the reaction (monitored by TLC). The mixture was then cooled to r.t. and the yellow solid was collected by filtration. The filtrate of ionic liquid [bmim$^+$][BF_4^-] and H_2O was then recycled for later use. The crude product was washed with H_2O and purified by recrystallization from 95% EtOH; this gave **6a**–**g**.

2-{[4,4-Dimethyl-2-(4-methylanilino)-6-oxocyclohex-1-enyl](phenyl)methyl}-3-hydroxy-5,5-dimethylcyclohex-2-enone (6a)

Pale yellow crystals; mp 209–211 °C.

IR (KBr): 3256, 3171, 3060, 2956, 2869, 1643, 1575, 1512, 1477, 1451, 1424, 1362, 1301, 1278, 1261, 1222, 1177, 1143, 1122, 1022, 1000, 921, 840, 803, 744, 697 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.93 (s, 3 H, CH_3), 1.06 (s, 6 H, 2 CH_3), 1.11 (s, 3 H, CH_3), 2.16 (d, *J* = 16.8 Hz, 1 H, CH), 2.28–2.42 (m, 9 H, CH_3 + 6 CH), 2.64 (d, *J* = 17.6 Hz, 1 H, CH), 5.59 (s, 1 H, CH), 6.96–7.26 (m, 9 H, ArH), 10.03 (s, 1 H, NH), 13.21 (s, 1 H, OH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 21.0, 28.0, 28.27, 28.31, 28.6, 31.77, 31.83, 32.7, 33.5, 41.2, 49.5, 111.1, 114.1, 114.9, 125.0, 125.5, 126.9, 127.1, 128.3, 130.3, 135.8, 136.0, 140.6, 196.4, 200.3.

Anal. Calcd for $C_{30}H_{35}NO_3$: C, 78.74; H, 7.71; N, 3.06. Found: C, 78.60; H, 7.80; N, 3.18.

2-{(2-Chlorophenyl)[4,4-dimethyl-2-(4-methylanilino)-6-oxocyclohex-1-enyl]methyl}-3-hydroxy-5,5-dimethylcyclohex-2-enone (6b)

Pale yellow crystals; mp 171–173 °C.

IR (KBr): 3255, 3169, 3029, 2955, 2925, 2869, 1630, 1600, 1511, 1467, 1408, 1326, 1248, 1151, 1120, 1101, 1035, 914, 830, 811, 765, 746, 718 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.91 (s, 3 H, CH_3), 0.97 (s, 3 H, CH_3), 1.01 (s, 6 H, 2 CH_3), 2.09 (d, *J* = 16.4 Hz, 1 H, CH), 2.19 (d, *J* = 16.0 Hz, 2 H, 2 CH), 2.26–2.46 (m, 8 H, CH_3 + 5 CH), 5.74 (s, 1 H, CH), 6.98 (d, *J* = 8.0 Hz, 2 H, ArH), 7.14–7.26 (m, 4 H, ArH), 7.30–7.33 (m, 2 H, ArH), 9.00 (s, 1 H, NH), 12.28 (s, 1 H, OH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.9, 21.3, 26.5, 28.0, 28.2 29.8, 32.0, 32.2, 32.5, 33.8, 42.0, 49.9, 111.1, 112.2, 125.1, 126.8, 127.8, 129.8, 129.9, 130.0, 130.3, 133.1, 136.4, 139.4, 143.6, 151.5, 195.3, 200.3.

Anal. Calcd for $C_{30}H_{34}ClNO_3$: C, 73.23; H, 6.96; N, 2.85. Found: C, 73.18; H, 7.07; N, 2.90.

2-{(3-Chlorophenyl)[4,4-dimethyl-2-(4-methylanilino)-6-oxocyclohex-1-enyl]methyl}-3-hydroxy-5,5-dimethylcyclohex-2-enone (6c)

Pale yellow crystals; mp 217–218 °C.

IR (KBr): 3256, 3178, 3061, 2953, 2907, 2867, 1680, 1629, 1582, 1512, 1475, 1421, 1394, 1327, 1313, 1272, 1154, 1113, 1039, 943, 922, 821, 790, 744 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.92 (s, 3 H, CH_3), 1.05 (s, 6 H, 2 CH_3), 1.10 (s, 3 H, CH_3), 2.16 (d, *J* = 16.0 Hz, 1 H, CH), 2.28–2.43 (m, 9 H, CH_3 + 6 CH), 2.63 (d, *J* = 17.6 Hz, 1 H, CH), 5.60 (s, 1 H, CH), 7.05–7.09 (m, 4 H, ArH), 7.17–7.30 (m, 4 H, ArH), 9.97 (s, 1 H, NH), 13.11 (s, 1 H, OH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 21.0, 27.87, 27.94, 28.2, 28.5, 31.2, 31.9, 32.7, 33.4, 44.1, 49.5, 110.5, 113.7, 114.4, 125.1, 125.6, 125.8, 127.1, 130.1, 130.3, 133.2, 135.9, 136.0, 143.6, 151.2, 196.2, 200.3.

Anal. Calcd for $C_{30}H_{34}ClNO_3$: C, 73.23; H, 6.96; N, 2.85. Found: C, 73.11; H, 7.00; N, 2.98.

2-{(4-Chlorophenyl)[4,4-dimethyl-2-(4-methylanilino)-6-oxocyclohex-1-enyl]methyl}-3-hydroxy-5,5-dimethylcyclohex-2-enone (6d)

Pale yellow crystals; mp 147–148 °C.

IR (KBr): 3252, 3122, 3029, 2956, 2868, 1678, 1638, 1579, 1513, 1488, 1467, 1400, 1330, 1312, 1276, 1222, 1161, 1094, 1037, 1011, 942, 921, 863, 830, 818 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.91 (s, 3 H, CH_3), 1.04 (s, 6 H, 2 CH_3), 1.10 (s, 3 H, CH_3), 2.15 (d, *J* = 16.4 Hz, 1 H, CH), 2.28–2.43 (m, 9 H, CH_3 + 6 CH), 2.63 (d, *J* = 16.4 Hz, 1 H, CH), 5.57 (s, 1 H, CH), 7.06 (d, *J* = 8.0 Hz, 2 H, ArH), 7.10 (d, *J* = 8.4 Hz, 2 H, ArH), 7.23 (d, *J* = 8.0 Hz, 2 H, ArH), 7.29 (d, *J* = 8.4 Hz, 2 H, ArH), 9.99 (s, 1 H, NH), 13.14 (s, 1 H, OH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 21.0, 21.3, 26.6, 28.0, 28.5, 29.7, 31.9, 32.4, 32.7, 33.2, 41.4, 49.9, 113.0, 125.0, 128.2, 128.3, 129.0, 129.9, 130.1, 130.3, 135.9, 136.0, 145.7, 151.2, 195.6, 200.3.

Anal. Calcd for $C_{30}H_{34}ClNO_3$: C, 73.23; H, 6.96; N, 2.85. Found: C, 73.10; H, 6.98; N, 3.01.

2-{[4,4-Dimethyl-2-(4-methylanilino)-6-oxocyclohex-1-enyl](4-methoxyphenyl)methyl}-3-hydroxy-5,5-dimethylcyclohex-2-enone (6e)

Pale yellow crystals; mp 144–146 °C.

IR (KBr): 3257, 3119, 3029, 2955, 2924, 2868, 2830, 1636, 1517, 1509, 1465, 1420, 1330, 1276, 1246, 1162, 1125, 1038, 893, 835, 820, 750 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.93 (s, 3 H, CH_3), 1.05 (s, 6 H, 2 CH_3), 1.11 (s, 3 H, CH_3), 2.15 (d, *J* = 16.0 Hz, 1 H, CH), 2.27–2.41 (m, 9 H, CH_3 + 6 CH), 2.62 (d, *J* = 16.0 Hz, 1 H, CH), 3.71 (s, 3 H, CH_3O), 5.53 (s, 1 H, CH), 6.81 (d, *J* = 8.0 Hz, 2 H, ArH), 6.99–7.06 (m, 4 H, ArH), 7.23 (d, *J* = 8.4 Hz, 2 H, ArH), 10.05 (s, 1 H, NH), 13.27 (s, 1 H, OH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 21.0, 21.2, 27.9, 28.0, 28.3, 28.4, 31.8, 32.4, 32.6, 32.7, 44.2, 49.5, 55.4, 113.7, 113.8, 123.7, 125.0, 128.1, 128.9, 130.1, 130.3, 135.8, 136.1, 150.7, 157.3, 196.3, 200.4.

Anal. Calcd for $C_{31}H_{37}NO_4$: C, 76.36; H, 7.65; N, 2.87. Found: C, 76.29; H, 7.80; N, 2.99.

2-{[4,4-Dimethyl-2-(4-methylanilino)-6-oxocyclohex-1-enyl](3-nitrophenyl)methyl}-3-hydroxy-5,5-dimethylcyclohex-2-enone (6f)

Pale yellow crystals; mp 190–192 °C.

IR (KBr): 3250, 3091, 3027, 2956, 2868, 1681, 1629, 1585, 1525, 1475, 1396, 1306, 1282, 1238, 1156, 1096, 1040, 942, 924, 804, 759, 732 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.95 (s, 3 H, CH_3), 1.08 (s, 6 H, 2 CH_3), 1.13 (s, 3 H, CH_3), 2.20 (d, *J* = 16.8 Hz, 1 H, CH), 2.28–2.48 (m, 9 H, CH_3 + 6 CH), 2.62 (d, *J* = 17.2 Hz, 1 H, CH), 5.73 (s, 1 H, CH), 7.08 (d, *J* = 8.0 Hz, 2 H, ArH), 7.24 (d, *J* = 8.0 Hz, 2 H, ArH), 7.53–7.59 (m, 3 H, ArH), 8.01 (d, *J* = 7.6 Hz, 1 H, ArH), 10.01 (s, 1 H, NH), 13.15 (s, 1 H, OH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.8, 21.1, 26.3, 27.9, 28.0, 29.5, 31.6, 32.3, 32.4, 32.7, 41.2, 49.6, 112.4, 121.3, 122.3, 125.0, 130.0, 130.1, 134.5, 135.6, 135.8, 143.3, 147.7, 148.0, 148.6, 151.6, 195.5, 195.9.

Anal. Calcd for $C_{30}H_{34}N_2O_5$: C, 71.69; H, 6.82; N, 5.57. Found: C, 71.50; H, 6.95; N, 5.60.

2-{[4,4-Dimethyl-2-(4-methylanilino)-6-oxocyclohex-1-enyl](4-fluorophenyl)methyl}-3-hydroxy-5,5-dimethylcyclohex-2-enone (6g)
Pale yellow crystals; mp 170–172 °C.

IR (KBr): 3250, 3060, 2958, 2865, 1680, 1589, 1504, 1470, 1395, 1327, 1271, 1227, 1155, 1115, 1038, 942, 871, 819, 800, 735 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.91 (s, 3 H, CH_3), 1.04 (s, 6 H, 2 CH_3), 1.10 (s, 3 H, CH_3), 2.15 (d, J = 16.0 Hz, 1 H, CH), 2.29–2.42 (m, 9 H, CH_3 + 6 CH), 2.64 (d, J = 17.6 Hz, 1 H, CH), 5.56 (s, 1 H, CH), 7.04–7.12 (m, 6 H, ArH), 7.23 (d, J = 8.0 Hz, 2 H, ArH), 9.99 (s, 1 H, NH), 13.18 (s, 1 H, OH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.9, 27.9, 28.0, 28.4, 28.6, 31.8, 32.4, 32.7, 33.0, 44.2, 49.5, 114.2, 114.8, 115.1, 123.7, 125.0, 128.75, 128.83, 130.3, 135.8, 136.0, 139.4, 151.1, 159.4, 161.8, 196.3, 200.3.

Anal. Calcd for $C_{30}H_{34}FNO_3$: C, 75.76; H, 7.21; N, 2.95. Found: C, 75.62; H, 7.25; N, 3.10.

9,10-Diarylacridine-1,8-diones 4aa–ag; General Procedure
A mixture of the appropriate aniline derivative **6** (1.0 mmol) in the ionic liquid medium [bmim$^+$][BF_4^-] (5 mL) was stirred at 90 °C for several hours to complete the reaction (monitored by TLC). The mixture was then cooled to r.t. and the yellow solid was collected by filtration. The filtrate of the ionic liquid [bmim$^+$][BF_4^-] was then recovered for reuse by drying at 80 °C several hours in vacuo. The crude product was washed with H_2O and purified by recrystallization from DMF–H_2O; this gave **4aa–ag**.

3,3,6,6-Tetramethyl-9-phenyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4aa)
Pale yellow crystals; mp 262–263 °C (Lit.[15] 262–263 °C).

9-(2-Chlorophenyl)-3,3,6,6-tetramethyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4ab)
Pale yellow crystals; mp 286–288 °C (Lit.[15] 285–287 °C).

9-(3-Chlorophenyl)-3,3,6,6-tetramethyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4ac)
Pale yellow crystals; mp >300 °C (Lit.[16] 315–317 °C).

9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4ad)
Pale yellow crystals; mp 284–286 °C (Lit.[15] 282–283 °C).

9-(4-Methoxyphenyl)-3,3,6,6-tetramethyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4ae)
Pale yellow crystals; mp 274–276 °C (Lit.[15] 273–274 °C).

3,3,6,6-Tetramethyl-9-(3-nitrophenyl)-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4af)
Pale yellow crystals; mp 285–286 °C (Lit.[16] 285–287 °C).

9-(4-Fluorophenyl)-3,3,6,6-tetramethyl-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4ag)
Pale yellow crystals; mp 259–261 °C.

IR (KBr): 3060, 2957, 2869, 1641, 1575, 1511, 1643, 1410, 1361, 1300, 1280, 1262, 1177, 1145, 1121, 999, 921, 843, 812, 715 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.71 (s, 6 H, 2 CH_3), 0.87 (s, 6 H, 2 CH_3), 1.77 (d, J = 17.6 Hz, 2 H, 2 CH), 2.00 (d, J = 16.0 Hz, 2 H, 2 CH), 2.17–2.22 (m, 4 H, 4 CH), 2.42 (s, 3 H, CH_3), 5.03 (s, 1 H, CH), 7.06 (d, J = 8.0 Hz, 2 H, ArH), 7.30–7.33 (m, 4 H, ArH), 7.41 (d, J = 7.6 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 20.9, 26.5, 28.4, 32.4, 32.7, 42.5, 50.6, 113.2, 114.9, 115.1, 123.7, 129.6, 129.7, 130.1, 134.2, 136.9, 139.4, 151.1, 161.0, 195.6.

Anal. Calcd for $C_{30}H_{32}FNO_2$: C, 78.75; H, 7.05; N, 3.06. Found: C, 78.66; H, 7.25; N, 3.22.

Indenoquinoline 8a via a 2-Benzylideneindene-1,3-dione Intermediate; Typical Procedure
A mixture of 4-bromobenzaldehyde (0.35 g, 2.0 mmol) and indene-1,3-dione (**7**; 0.30 g, 2.0 mmol) in the ionic liquid medium [bmim$^+$][BF_4^-] (5 mL) was stirred at 90 °C for 2 hours to complete the reaction (monitored by TLC). The mixture was then cooled to r.t. and the yellow solid was collected by filtration. The filtrate of the ionic liquid [bmim$^+$][BF_4^-] was then recovered for reuse by drying at 80 °C for several hours in vacuo. The crude yellow product was washed with H_2O and purified by recrystallization from 95% EtOH; this gave 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione. A mixture of this intermediate (0.33 g, 1.0 mmol) and 5,5-dimethyl-3-(4-methylanilino)cyclohex-2-enone (**2a**, 1.0 mmol) in [bmim$^+$][BF_4^-] (5 mL) was stirred at 90 °C for 2 h to complete the reaction (monitored by TLC). The mixture was then cooled to r.t., the red solid was collected by filtration, and the filtrate of the ionic liquid [bmim$^+$][BF_4^-] was then recovered for reuse by drying at 80 °C for several hours in vacuo. The crude red product was washed with H_2O, purified by recrystallization from DMF–H_2O, and dried at 80 °C for several hours in vacuo; this gave **8a**; yield: 97%.

2-(4-Bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione (8a)
Pale yellow crystals; mp 179–181 °C.

IR (KBr): 3054, 1690, 1632, 1593, 1483, 842, 738, 707 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 7.81 (d, J = 8.4 Hz, 2 H, ArH), 7.85 (s, 1 H, ArH), 7.97–8.04 (m, 4 H, ArH), 8.45 (d, J = 8.4 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 123.6, 129.3, 130.2, 132.1, 135.9, 136.5, 136.6, 138.4, 140.0, 142.4, 144.2, 189.1, 189.7.

Anal. Calcd for $C_{16}H_9BrO_3$: C, 61.37; H, 2.90; Found: C, 61.30; H, 2.91.

Indenoquinolines 8a–i by Three-Component Reactions between Benzaldehydes 1, 3-Anilino-5,5-dimethylcyclohex-2-enones 2, and Indene-1,3-dione (7); General Procedure
A mixture of benzaldehyde **1** (1.0 mmol), 3-anilino-5,5-dimethylcyclohex-2-enone **2** (1.0 mmol), indene-1,3-dione (**7**; 0.15 g, 1.0 mmol) and the ionic liquid medium [bmim$^+$][BF_4^-] (5 mL) was stirred at 90 °C for several hours to complete the reaction (monitored by TLC). The mixture was then cooled to r.t., and the red solid was collected by filtration. The filtrate of the ionic liquid [bmim$^+$][BF_4^-] was then recovered for reuse by drying at 80 °C for several hours in vacuo. The crude red product was washed with H_2O, purified by recrystallization from DMF–H_2O, and dried at 80 °C for several hours in vacuo; this gave **8a–i**.

10-(4-Bromophenyl)-7,7-dimethyl-5-(4-tolyl)-6,7,8,10-tetrahydro-5*H*-indeno[1,2-*b*]quinoline-9,11-dione (8a)
Red crystals; mp 275–277 °C.

IR (KBr): 3050, 2960, 2887, 1693, 1642, 1588, 1557, 1511, 1485, 1455, 1396, 1366, 1255, 1223, 1170, 1140, 1101, 1057, 1012, 889, 842, 763, 728, 710 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.80 (s, 3 H, CH_3), 0.94 (s, 3 H, CH_3), 1.99 (d, *J* = 17.6 Hz, 1 H, CH), 2.05 (d, *J* = 16.0 Hz, 1 H, CH), 2.27 (d, *J* = 16.0 Hz, 1 H, CH), 2.39 (d, *J* = 17.6 Hz, 1 H, CH), 2.50 (s, 3 H, CH_3), 4.83 (s, 1 H, CH), 5.21 (d, *J* = 7.6 Hz, 1 H, ArH), 7.01–7.04 (m, 1 H, ArH), 7.19–7.25 (m, 2 H, ArH), 7.33 (d, *J* = 8.4 Hz, 2 H, ArH), 7.45–7.50 (m, 4 H, ArH), 7.54–7.61 (m, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 21.0, 26.2, 29.4, 32.0, 33.1, 40.6, 49.6, 111.2, 114.5, 120.9, 121.0, 128.2, 129.9, 130.0, 130.5, 130.9, 132.0, 133.0, 135.6, 136.9, 140.3, 144.7, 151.5, 154.8, 191.1, 195.9.

Anal. Calcd for $C_{31}H_{26}BrNO_2$: C, 71.00; H, 5.00; N, 2.67. Found: C, 70.79; H, 5.09; N, 2.70.

10-(4-Fluorophenyl)-7,7-dimethyl-5-(4-tolyl)-6,7,8,10-tetrahydro-5*H*-indeno[1,2-*b*]quinoline-9,11-dione (8b)
Red crystals; mp 218–220 °C.

IR (KBr): 3050, 2959, 2892, 2838, 1673, 1633, 1601, 1588, 1558, 1498, 1455, 1398, 1362, 1255, 1222, 1191, 1158, 1139, 1102, 1091, 1061, 1017, 888, 852, 766, 705 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.79 (s, 3 H, CH_3), 0.94 (s, 3 H, CH_3), 1.99 (d, *J* = 17.6 Hz, 1 H, CH), 2.02 (d, *J* = 16.0 Hz, 1 H, CH), 2.26 (d, *J* = 16.0 Hz, 1 H, CH), 2.52 (d, *J* = 17.6 Hz, 1 H, CH), 2.50 (s, 3 H, CH_3), 4.85 (s, 1 H, CH), 5.20 (d, *J* = 7.6 Hz, 1 H, ArH), 7.02–7.11 (m, 3 H, ArH), 7.16–7.24 (m, 2 H, ArH), 7.37–7.41 (m, 2 H, ArH), 7.48 (d, *J* = 7.6 Hz, 2 H, ArH), 7.53–7.61 (m, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 21.1, 26.2, 29.5, 32.0, 32.8, 40.3, 50.0, 111.5, 114.7, 114.8, 115.0, 120.9, 121.0, 129.7, 129.8, 129.9, 132.0, 133.0, 135.6, 137.0, 140.3, 141.98, 142.01, 151.3, 154.6, 159.7, 162.5, 191.2, 195.6.

Anal. Calcd for $C_{31}H_{26}FNO_2$: C, 80.32; H, 5.65; N, 3.02. Found: C, 80.15; H, 5.68; N, 3.12.

5,10-Bis(4-bromophenyl)-7,7-dimethyl-5-(4-tolyl)-6,7,8,10-tetrahydro-5*H*-indeno[1,2-*b*]quinoline-9,11-dione (8c)
Red crystals; mp 271–273 °C.

IR (KBr): 3089, 2959, 2883, 1692, 1645, 1608, 1590, 1559, 1487, 1455, 1395, 1365, 1301, 1254, 1223, 1191, 1169, 1141, 1100, 1070, 1010, 976, 888, 841, 791, 763, 705 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.80 (s, 3 H, CH_3), 0.94 (s, 3 H, CH_3), 1.98 (d, *J* = 17.6 Hz, 1 H, CH), 2.05 (d, *J* = 16.0 Hz, 1 H, CH), 2.26 (d, *J* = 16.0 Hz, 1 H, CH), 2.38 (d, *J* = 17.6 Hz, 1 H, CH), 4.82 (s, 1 H, CH), 5.26 (d, *J* = 7.6 Hz, 1 H, ArH), 7.07–7.12 (m, 1 H, ArH), 7.19–7.26 (m, 2 H, ArH), 7.34 (d, *J* = 8.0 Hz, 2 H, ArH), 7.45 (d, *J* = 8.0 Hz, 2 H, ArH), 7.72 (b, 2 H, ArH), 7.90 (d, *J* = 8.4 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.5, 29.7, 32.3, 34.5, 40.6, 49.8, 111.9, 114.8, 119.8, 121.1, 121.7, 124.1, 130.3, 130.7, 131.4, 132.5, 132.6, 133.1, 137.1, 137.8, 145.3, 151.4, 154.6, 191.4, 195.9.

Anal. Calcd for $C_{30}H_{23}Br_2NO_2$: C, 61.14; H, 3.93; N, 2.38. Found: C, 61.00; H, 3.98; N, 2.49.

5-(4-Bromophenyl)-10-(4-fluorophenyl)-7,7-dimethyl-6,7,8,10-tetrahydro-5*H*-indeno[1,2-*b*]quinoline-9,11-dione (8d)
Red crystals; mp 280–282 °C.

IR (KBr): 3047, 2955, 2891, 1686, 1650, 1636, 1592, 1566, 1503, 1488, 1453, 1398, 1369, 1312, 1259, 1225, 1191, 1170, 1155, 1101, 1012, 978, 940, 888, 852, 764, 734, 705 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.80 (s, 3 H, CH_3), 0.94 (s, 3 H, CH_3), 1.98 (d, *J* = 17.6 Hz, 1 H, CH), 2.05 (d, *J* = 16.0 Hz, 1 H, CH), 2.26 (d, *J* = 16.0 Hz, 1 H, CH), 2.38 (d, *J* = 17.6 Hz, 1 H, CH), 4.84 (s, 1 H, CH), 5.26 (d, *J* = 7.6 Hz, 1 H, ArH), 7.06–7.10 (m, 3 H, ArH), 7.19–7.26 (m, 2 H, ArH), 7.38–7.42 (m, 2 H, ArH), 7.71 (b, 2 H, ArH), 7.90 (d, *J* = 8.8 Hz, 2 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.1, 30.9, 32.0, 32.8, 40.2, 49.6, 111.8, 114.8, 115.0, 120.7, 121.2, 123.8, 129.8, 129.9, 130.0, 132.2, 132.3, 132.8, 133.2, 136.8, 137.5, 141.8, 150.8, 154.2, 159.7, 162.1, 191.2, 195.6.

Anal. Calcd for $C_{30}H_{23}BrFNO_2$: C, 68.19; H, 4.39; N, 2.65. Found: C, 68.10; H, 4.45; N, 2.70.

5-(4-Bromophenyl)-10-(2,4-dichlorophenyl)-7,7-dimethyl-6,7,8,10-tetrahydro-5*H*-indeno[1,2-*b*]quinoline-9,11-dione (8e)
Red crystals; mp 288–290 °C.

IR (KBr): 3062, 2954, 2871, 1683, 1636, 1591, 1582, 1566, 1489, 1470, 1396, 1368, 1301, 1257, 1226, 1193, 1169, 1154, 1101, 1063, 1012, 889, 860, 765, 734, 703 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.83 (s, 3 H, CH_3), 0.94 (s, 3 H, CH_3), 1.98 (d, *J* = 17.6 Hz, 1 H, CH), 2.01 (d, *J* = 16.0 Hz, 1 H, CH), 2.24 (d, *J* = 16.0 Hz, 1 H, CH), 2.33 (d, *J* = 17.6 Hz, 1 H, CH), 5.20 (s, 1 H, CH), 5.23 (d, *J* = 7.6 Hz, 1 H, ArH), 7.07–7.11 (m, 1 H, ArH), 7.21–7.23 (m, 2 H, ArH), 7.33 (dd, *J* = 8.4, 1.6 Hz, 1 H, ArH), 7.47 (d, *J* = 1.6 Hz, 1 H, ArH), 7.53 (d, *J* = 8.4 Hz, 1 H, ArH), 7.67–7.75 (m, 2 H, ArH), 7.89–7.92 (m, 2 H, ArH).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 26.9, 29.7, 32.2, 33.7, 41.1, 49.9, 114.3, 120.7, 121.5, 124.5, 127.0, 129.7, 129.9, 131.5, 131.7, 132.8, 133.1, 133.2, 133.4, 134.1, 134.5, 136.8, 137.7, 139.9, 151.2, 154.7, 191.6, 195.9.

Anal. Calcd for $C_{30}H_{22}BrCl_2NO_2$: C, 62.20; H, 3.83; N, 2.42. Found: C, 62.11; H, 3.85; N, 2.58.

10-(3-Chlorophenyl)-7,7-dimethyl-5-(1-naphthyl)-6,7,8,10-tetrahydro-5*H*-indeno[1,2-*b*]quinoline-9,11-dione (8f)
Red crystals; mp 225–226 °C.

IR (KBr): 3054, 2960, 2871, 1689, 1652, 1635, 1590, 1563, 1468, 1455, 11391, 1363, 1301, 1260, 1226, 1191, 1171, 1134, 1004, 890, 728, 730, 685 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.79 (s, 3 H, CH_3), 0.80 (s, 3 H, CH_3), 1.94 (d, *J* = 17.4 Hz, 1 H, CH), 2.11 (d, *J* = 16.0 Hz, 1 H, CH), 2.25 (d, *J* = 16.4 Hz, 2 H, 2 CH), 4.68 (d, *J* = 7.6 Hz, 1 H, ArH), 4.98 (s, 1 H, CH), 6.72–6.76 (m, 1 H, ArH), 6.76–7.45 (m, 6 H, ArH), 7.65–7.70 (m, 2 H, ArH), 7.80–7.85 (m, 2 H, ArH), 8.03 (d, *J* = 6.4 Hz, 1 H, ArH), 8.20 (d, *J* = 7.2 Hz, 1 H, ArH), 8.34 (d, *J* = 8.4 Hz, 1 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 27.3, 28.6, 32.5, 34.2, 40.6, 50.0, 111.5, 114.8, 120.7, 121.4, 122.4, 126.6, 126.7, 127.2, 128.0, 128.4, 128.9, 129.2, 129.4, 130.2, 130.6, 131.5, 131.8, 132.2, 133.1, 133.2, 134.1, 134.6, 136.7, 148.4, 152.1, 155.2, 191.5, 196.0.

Anal. Calcd for $C_{34}H_{26}ClNO_2$: C, 79.14; H, 5.08; N, 2.71. Found: C, 78.95; H, 5.21; N, 2.70.

10-(2-Chlorophenyl)-7,7-dimethyl-5-(1-naphthyl)-6,7,8,10-tetrahydro-5*H*-indeno[1,2-*b*]quinoline-9,11-dione (8g)
Red crystals; mp 260–263 °C.

IR (KBr): 3056, 3013, 2952, 2868, 1695, 1633, 1606, 1560, 1507, 1467, 1457, 1395, 1363, 1261, 1227, 1192, 1172, 1132, 1037, 1005, 890, 782, 766, 733, 700 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 0.79 (s, 3 H, CH_3), 0.82 (s, 3 H, CH_3), 1.93 (d, *J* = 17.4 Hz, 1 H, CH), 2.03 (d, *J* = 16.4 Hz, 1 H, CH), 2.11 (d, *J* = 17.2 Hz, 1 H, CH), 2.22 (d, *J* = 16.4 Hz, 1 H, 1 CH), 4.66 (d, *J* = 7.6 Hz, 1 H, ArH), 5.38 (s, 1 H, CH), 6.72–6.76 (m, 1 H, ArH), 7.05–7.35 (m, 5 H, ArH), 7.61 (dd, *J* = 7.6, 1.2 Hz, 1 H, ArH), 7.66–7.69 (m, 2 H, ArH), 7.80–7.91 (m, 2 H, ArH), 8.01 (d, *J* = 7.2 Hz, 2 H, ArH), 8.19–8.22 (m, 1 H, ArH), 8.34 (d, *J* = 8.4 Hz, 1 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 27.2, 28.7, 32.4, 36.3, 40.6, 50.1, 115.0, 120.6, 121.2, 122.3, 126.6, 127.6, 128.0, 128.3, 129.1,

129.2, 129.4, 129.7, 130.2, 131.4, 131.9, 132.0, 132.1, 133.1, 133.2, 134.1, 134.7, 136.7, 152.3, 155.4, 162.8, 191.2, 195.7.

Anal. Calcd for $C_{34}H_{26}ClNO_2$: C, 79.14; H, 5.08; N, 2.71. Found: C, 78.90; H, 5.24; N, 2.75.

10-(4-Methoxyphenyl)-7,7-dimethyl-5-(1-naphthyl)-6,7,8,10-tetrahydro-5*H*-indeno[1,2-*b*]quinoline-9,11-dione (8h)
Red crystals; mp 256–258 °C.

IR (KBr): 3056, 2995, 2960, 2932, 2877, 2832, 2685, 1654, 1633, 1589, 1562, 1508, 1455, 1392, 1364, 1300, 1251, 1226, 1192, 1170, 1134, 1037, 1004, 889, 784, 763, 708 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.80 (s, 6 H, 2 CH_3), 1.95 (d, *J* = 16.8 Hz, 1 H, CH), 2.07 (d, *J* = 16.0 Hz, 1 H, CH), 2.18 (d, *J* = 16.8 Hz, 1 H, CH), 2.24 (d, *J* = 16.0 Hz, 1 H, 1 CH), 3.74 (s, 3 H, CH_3O), 4.66 (d, *J* = 7.6 Hz, 1 H, ArH), 4.91 (s, 1 H, CH), 6.70–6.74 (m, 1 H, ArH), 6.86 (d, *J* = 8.4 Hz, 2 H, ArH), 7.03–7.19 (m, 2 H, ArH), 7.36 (d, *J* = 8.4 Hz, 2 H, ArH), 7.65–7.68 (m, 4 H, ArH), 7.99 (d, *J* = 7.2 Hz, 2 H, ArH), 8.19–8.21 (m, 1 H, ArH), 8.33 (d, *J* = 8.4 Hz, 1 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 26.9, 28.5, 32.2, 32.7, 38.9, 49.8, 55.1, 112.3, 113.7, 115.4, 120.2, 121.0, 122.1, 126.3, 127.7, 128.5, 128.9, 129.1, 129.7, 131.1, 131.6, 131.8, 133.0, 133.8, 134.5, 136.6, 138.2, 151.0, 154.5, 157.9, 191.3, 195.7.

Anal. Calcd for $C_{35}H_{29}NO_3$: C, 82.17; H, 5.71; N, 2.74. Found: C, 82.00; H, 5.92; N, 2.81.

10-(4-Fluorophenyl)-7,7-dimethyl-5-(1-naphthyl)-6,7,8,10-tetrahydro-5*H*-indeno[1,2-*b*]quinoline-9,11-dione (8i)
Red crystals; mp 267–270 °C.

IR (KBr): 3059, 2954, 2868, 1683, 1652, 1631, 1591, 1563, 1505, 1455, 1393, 1363, 1263, 1226, 1192, 1156, 1128, 1005, 889, 864, 786, 768, 707 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 0.78 (s, 3 H, CH_3), 0.79 (s, 3 H, CH_3), 1.95 (d, *J* = 17.2 Hz, 1 H, CH), 2.09 (d, *J* = 16.4 Hz, 1 H, CH), 2.18 (d, *J* = 17.2 Hz, 1 H, CH), 2.24 (d, *J* = 16.4 Hz, 1 H, 1 CH), 4.67 (d, *J* = 7.6 Hz, 1 H, ArH), 4.98 (s, 1 H, CH), 6.71–6.75 (m, 1 H, ArH), 7.04–7.22 (m, 4 H, ArH), 7.47–7.50 (m, 2 H, ArH), 7.65–7.71 (m, 2 H, ArH), 7.79–7.85 (m, 2 H, ArH), 8.02 (d, *J* = 7.2 Hz, 2 H, ArH), 8.19–8.21 (m, 1 H, ArH), 8.34 (d, *J* = 8.0 Hz, 1 H, ArH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 28.7, 29.7, 32.5, 33.4, 40.6, 50.1, 112.0, 115.1, 115.3, 120.6, 121.3, 122.4, 126.6, 128.0, 128.9, 129.2, 129.4, 130.1, 130.2, 130.3, 131.4, 131.9, 132.2, 133.2, 134.1, 134.7, 136.8, 142.4, 151.7, 155.1, 160.0, 162.4, 191.5, 196.0.

Anal. Calcd for $C_{34}H_{26}FNO_2$: C, 81.74; H, 5.25; N, 2.80. Found: C, 81.68; H, 5.30; N, 2.88.

Acknowledgment

We are grateful to the Natural Science Foundation (04KJB150139) of Jiangsu Education Committee for financial support.

References

(1) (a) Welton, T. *Chem. Rev.* **1999**, *99*, 2071. (b) Wasserscheid, P.; Keim, W. *Angew. Chem. Int. Ed.* **2000**, *39*, 3773. (c) Sheldon, R. *Chem. Commun.* **2001**, 2399. (d) Wilkes, J. S. *Green Chem.* **2002**, *4*, 73.

(2) (a) da Silveira Neto, B. A.; Ebeling, G.; Goncalves, R. S.; Gozzo, F. C.; Eberlin, M. N.; Dupont, J. *Synthesis* **2004**, 1155. (b) Yeung, K. S.; Farkas, M. E.; Qiu, Z.; Yang, Z. *Tetrahedron Lett.* **2002**, *43*, 5793. (c) Fraga-Dubreuil, J.; Bazureau, J. P. *Tetrahedron* **2003**, *59*, 6121. (d) Yadav, J. S.; Reddy, B. V. S.; Basak, A. K.; Narsaiah, A. V. *Tetrahedron Lett.* **2003**, *44*, 1047. (e) Su, C.; Chen, Z. C.; Zheng, Q. G. *Synthesis* **2003**, 555. (f) Hakkou, H.; Jacques, J.; Eynde, V.; Hamelin, J.; Bazureau, J. P. *Synthesis* **2004**, 1793.

(3) (a) Janis, R. A.; Silver, P. J.; Triggle, D. J. *Adv. Drug Res.* **1987**, *16*, 309. (b) Shan, R.; Velazquez, C.; Knaus, E. E. *J. Med. Chem.* **2004**, *41*, 254. (c) Dulhunty, A. F.; Curtis, S. M.; Watson, S.; Cengia, L.; Marco, G. *J. Biol. Chem.* **2004**, *279*, 11853.

(4) Martin, N.; Quinteiro, M.; Seoane, C.; Mora, L.; Suarez, M.; Ockoa, E.; Morales, A. *J. Heterocycl. Chem.* **1995**, *51*, 235.

(5) (a) Wang, X. S.; Zhang, M. M.; Zeng, Z. S.; Shi, D. Q.; Tu, S. J.; Wei, X. Y.; Zong, Z. M. *Tetrahedron Lett.* **2005**, *46*, 7169. (b) Wang, X. S.; Shi, D. Q.; Zhang, Y. F.; Wang, S. H.; Tu, S. J. *Chin. J. Org. Chem.* **2004**, *24*, 430.

(6) Tu, S. J.; Miao, C. B.; Gao, Y.; Feng, Y. J.; Feng, J. C. *Chin. J. Chem.* **2002**, *20*, 703.

(7) Li, Y. L.; Zhang, M. M.; Wang, X. S.; Shi, D. Q.; Tu, S. J.; Wei, X. Y.; Zong, Z. M. *J. Chem. Res.* **2005**, 600.

(8) Mikata, Y.; Yokoyama, M.; Mogami, K.; Kato, M.; Okura, I.; Chikira, M.; Yano, S. *Inorg. Chim. Acta* **1998**, *279*, 51.

(9) Antonini, I.; Polucci, P.; Kelland, L. R.; Menta, E.; Pescalli, N.; Martelli, S. *J. Med. Chem.* **1999**, *42*, 2535.

(10) Gamage, S. A.; Spicer, J. A.; Atwell, G. J.; Finlay, G. J.; Baguley, B. C.; Denny, W. A. *J. Med. Chem.* **1999**, *42*, 2383.

(11) Eid, N. M.; Safwat, H. M.; Ramadan, M. A.; Shouman, S. A.; El-Esawy, A. A. *Bull. Fac. Pharm. (Cairo Univ.)* **1995**, *33*, 25.

(12) Gallo, S.; Atifi, S.; Mahamoud, A.; Santelli-Rouvier, C.; Wolfart, K.; Molnar, J.; Barbe, J. *Eur. J. Med. Chem.* **2003**, *38*, 19.

(13) Srivastava, A.; Nizamuddin, C. *Indian J. Heterocycl. Chem.* **2004**, *13*, 261.

(14) Ferencz, L.; Peter, M. *Farmacia* **2003**, *51*, 52.

(15) Wang, X. S.; Shi, D. Q.; Wang, S. H.; Tu, S. J. *Chin. J. Org. Chem.* **2003**, *23*, 1291.

(16) Jin, T. S.; Zhang, J. S.; Guo, T. T.; Wang, A. Q.; Li, T. S. *Synthesis* **2004**, 2001.

(17) (a) Chebanov, V. A.; Saraev, V. E.; Kobzar, K. M.; Desenko, S. M.; Orlov, V. D.; Gura, E. A. *Chem. Heterocycl. Compd.* **2004**, *40*, 475. (b) El-Sadek, M. I.; Al-Ashmawi, M. A.; El-Bermawy, M. A.; Mohamed, A. K.; Al-Sabbagh, O. I. *Zagazig, J. Pharm. Sci.* **1994**, *3*, 144.

(18) Abdel-Gawad, S. M.; El-Gaby, M. S. A.; Heiba, H. I.; Aly, H. M.; Ghorab, M. M. *J. Chin. Chem. Soc.* **2005**, *52*, 1227.

(19) Crystal structure data for **4r**: $C_{29}H_{31}NO_3$, *M* = 441.55, pale yellow block crystals, 0.50 × 0.47 × 0.26 mm^3, triclinic, space group *P*–1, *a* = 9.6382 (15), *b* = 11.5847 (15), *c* = 12.4722 (11) Å, α = 66.075 (9), β = 70.176 (9), γ = 85.800(13)°, *V* = 1194.1 (3) $Å^3$, *Z* = 2, D_c = 1.228 g·cm^{-3}, *F*(000) = 472, μ(Mo Kα) = 0.079 mm^{-1}. Intensity data were collected on a Rigaku Mercury diffractometer with graphite monochromated Mo-Kα radiation (λ = 0.71070 Å) using the ω scan mode with 3.07° < θ < 25.34°; 4332 unique reflections were measured and 3733 reflections with $I > 2\sigma(I)$ were used in the refinement. The structure was solved by direct methods and expanded using Fourier techniques. The final cycle of full-matrix least-squares technique refined to *R* = 0.0539 and *wR* = 0.1204.

(20) Shi, D. Q.; Zhuang, Q. Y.; Chen, J.; Wang, X. S.; Tu, S. J.; Hu, H. W. *Chin. J. Org. Chem.* **2003**, *23*, 694.

(21) Crystal structure data for **6a**: $C_{30}H_{35}NO_3$, M = 457.59, pale yellow block crystals, 0.20 × 0.15 × 0.10 mm^3, monoclinic, space group $P21/c$, a = 10.2337 (13), b = 17.077 (2), c = 15.2788 (19) Å, α = 90.00, β = 106.329 (3), γ = 90.00°, V = 2562.4 (5) Å^3, Z = 4, D_c = 1.186 g·cm^{-3}, F(000) = 984, μ(Mo Kα) = 0.076 mm^{-1}. Intensity data were collected on a CCD area diffractometer with graphite monochromated Mo-Kα radiation (λ = 0.71073Å) using the φ and ω scan modes with 2.07° < θ < 26.00°; 4985 unique reflections were measured and 1823 reflections with $I > 2\sigma(I)$ were used in the refinement. The structure was solved by direct methods and expanded using Fourier techniques. The final cycle of full-matrix least-squares technique refined to R = 0.0617 and wR = 0.1201.

(22) Crystal structure data for **8b**: $C_{34}H_{33}FN_2O_3$, M = 536.62, red block crystals, 0.40 × 0.21 × 0.20 mm^3, monoclinic, space group $P21/n$, a = 113.0147 (18), b = 9.5006 (12), c = 22.985 (3) Å, α = 90.00, β = 102.850 (3), γ = 90.00°, V = 2770.9 (6) Å^3, Z = 4, D_c = 1.286 g·cm^{-3}, F(000) = 1136, μ(Mo Kα) = 0.087 mm^{-1}. Intensity data were collected on a Rigaku Mercury diffractometer with graphite monochromated Mo-Kα radiation (λ = 0.71070Å) using the φ and ω scan modes with 3.03° < θ < 25.34°; 3830 unique reflections were measured and 1823 reflections with $I > 2\sigma(I)$ were used in the refinement. The structure was solved by direct methods and expanded using Fourier techniques. The final cycle of full-matrix least-squares technique refined to R = 0.0789 and wR = 0.1544.