An Efficient Synthesis of Polyhydroacridine Derivatives by the Three-component Reaction of Aldehydes, Amines and Dimedone in Ionic Liquid

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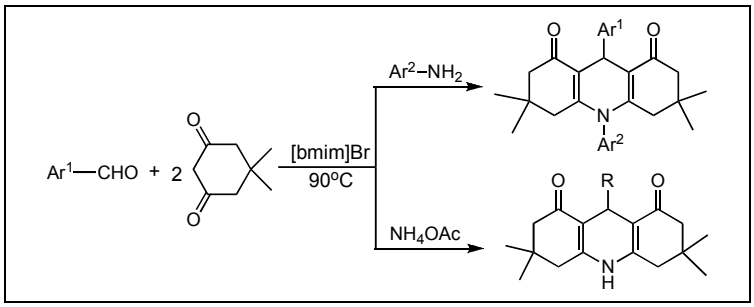

In this paper the preparation of 3,3,6,6-tetramethyl-9,10-diaryl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione derivatives from aldehydes, aromatic amines and 5,5-dimethyl-1,3-cyclohexanedione in 1-n-butyl-3-methylimidazolium bromide ([bmim]Br) is described. The structures of these compounds were characterized by elemental analysis, IR and ^{1}H NMR spectra and further confirmed by single crystal X-ray diffraction analysis.

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INTRODUCTION

Multi-component reactions (MCRs), in which multiple reactions are combined into one synthetic operation, have been used extensively to form carbon-carbon bonds in synthetic chemistry [1]. Such reactions offer a wide range of possibilities for the efficient construction of highly complex molecules in a single procedural step. Thus avoiding the complicate purification operations and allowing savings of both solvents and reagents. In the past decade there have been tremendous development in three- and four-component reactions and great efforts continue to be made to develop new MCRs [2-4].

Recently, the replacement of current chemical process with more environmentally benign alternatives is an increasingly attractive goal in organic synthesis. In this field, room temperature ionic liquid have been the subject of considerable current interest as environmentally benign reaction media in organic synthesis because of their unique properties of nonvolatility, nonflammability and recyclability, among others [5-7]. Numerous chemical reactions, such as hydrogenation [8-10], regioselective alkylation [11], Friedel-Crafts reactions [12-14], dimerization of alkenes [15], Diels-Alder reactions [16], Michael reactions [17], Cross-coupling reactions [18-20] and some enzymic reactions [21] can be carried out in ionic liquid.

1,4-Dihydropyridines (1,4-DHPs) are well-known compounds because of their biological activities [22-24]. Recently there have been many methods reported for the synthesis of tricyclic compounds containing 1,4-dihydropyridines, such as acridine derivatives, from aldehydes, dimedone and aromatic amines by traditional heating in organic solvents [25], or in water catalyzed by 4-dodecylbenzenesulfonic acid (DBSA) [26], or improved under microwave irradiation [27]. However, they were reacted in organic solvents or had low solubility in water and none of the acridine compounds contained N-aryl substituted with electron-withdrawing groups at the *para*-position was obtained. As a consequence of our interest in green synthesis [28-30], herein, we would like to report a highly efficient method for the synthesis of a series of polyhydroacridine derivatives by the three-component reaction of aldehydes, aromatic amines and dimedone in ionic liquid.

RESULTS AND DISCUSSION

When the three-components of aromatic aldehyde **1**, 5,5-dimethyl-1,3-cyclohexamedione **2** and aromatic amine **3** were treated in an ionic liquid 1-n-butyl-3-methylimidazolium bromide ([bmim]Br) at 90 °C for a

few hours (Scheme 1), the desired 3,3,6,6-tetramethyl-9,10-diaryl-1,2,3,4,5,6,7,8,9,10-decahydro-acridin-1,8-dione derivatives **4** were obtained in high yields (70% - 99%) (Table 1).

Scheme 1

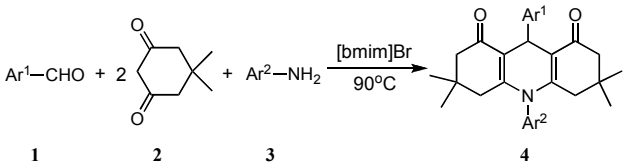


Table 1

Synthesis of **4** in [bmim]Br.

Product	Ar^1	Ar^2	Time (h)	Yield (%)
4a	4-BrC_6H_4	4-$CH_3OC_6H_4$	3	96
4b	4-$CH_3OC_6H_4$	4-$CH_3OC_6H_4$	2	99
4c	3,4-$OCH_2OC_6H_3$	4-$CH_3OC_6H_4$	4	92
4d	4-$CH_3C_6H_4$	4-$CH_3OC_6H_4$	2	98
4e	3,4-$(CH_3O)_2C_6H_3$	4-$CH_3OC_6H_4$	2.5	92
4f	4-$(CH_3)_2NC_6H_4$	4-$CH_3OC_6H_4$	4.5	80
4g	4-ClC_6H_4	4-$CH_3OC_6H_4$	2	96
4h	3,4-$Cl_2C_6H_3$	4-$CH_3OC_6H_4$	4	87
4i	4-HOC_6H_4	4-$CH_3OC_6H_4$	2.5	85
4j	4-BrC_6H_4	3-Cl-4-$CH_3C_6H_3$	3.5	87
4k	4-ClC_6H_4	3-Cl-4-$CH_3C_6H_3$	3.5	88
4l	4-$CH_3C_6H_4$	3-Cl-4-$CH_3C_6H_3$	3	89
4m	Thiophen-2-yl	4-$CH_3OC_6H_4$	4	79
4n	Pyrid-3-yl	4-$CH_3OC_6H_4$	4.5	83
4o	4-ClC_6H_4	C_6H_5	3.5	85
4p	4-ClC_6H_4	4-FC_6H_4	3.5	88
4q	4-$CH_3OC_6H_4$	4-ClC_6H_4	4	86
4r	4-$CH_3C_6H_4$	2-$CH_3C_6H_4$	4	70

As shown in Table 1, this method could be applied not only to aromatic aldehydes with either electron-withdrawing groups (such as halide groups) or electron-donating groups (such as alkyl, hydroxyl groups), but also to heterocyclic aldehydes. Furthermore, it was particularly noteworthy that the method could be applied to aromatic amine with either electron-donating groups (such as alkyl, alkoxyl groups) or electron-withdrawing groups (such as halide groups), which highlighted the wide scope of this three-component reaction. Therefore, we concluded that the electronic nature of the substituents of aldehydes and anilines has no significant effect on this reaction.

As expected, when the aromatic aldehydes **1** was replaced by dicarboxaldehydes **5**, another series of bis(decahydroacridine-1,8-dione) **6** were obtained under the same reaction conditions (Scheme 2). The results are summarized in Table 2.

Scheme 2

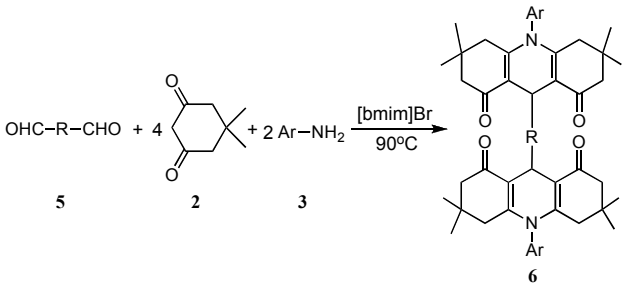


Table 2

Synthesis of **6** in [bmim]Br.

Product	R	Ar	Time (h)	Yield (%)
6a	1,3-C_6H_4	4-$CH_3OC_6H_4$	3.5	93
6b	1,3-C_6H_4	4-$CH_3C_6H_4$	2.5	90
6c	1,3-C_6H_4	C_6H_5	2.5	93
6d	1,4-C_6H_4	4-$CH_3C_6H_4$	3	87

To expand the reaction scope of aldehyde and dimedone with amine, we tried the reaction of **1** and **2** with ammonium acetate **7**, the desired products **8** were obtained in high yields (Scheme 3). The reaction time is shorter than that of the other methods [31-34]. Results are summarized in Table 3.

Scheme 3

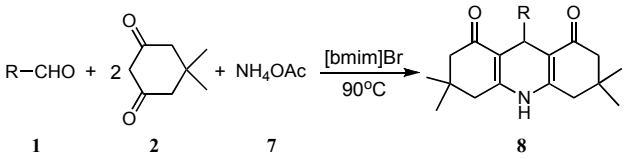


Table 3

Synthesis of **8** in [bmim]Br.

Product	R	Time (min)	Yield (%)	m. p. (lit. m. p.) (°C)
8a	4-$CH_3C_6H_4$	15	85	>300 (>300[35])
8b	4-$CH_3OC_6H_4$	15	90	272-274 (270-272 [36])
8c	4-BrC_6H_4	30	93	>300 (>300[35])
8d	3,4-$(CH_3O)_2C_6H_3$	30	92	261-263 (260-262 [37])
8e	4-$(CH_3)_2NC_6H_4$	30	90	269-271 (264-266 [36])
8f	4-HOC_6H_4	15	97	>300 (>300[35])
8g	3,4-$Cl_2C_6H_3$	50	92	
8h	2,4-$Cl_2C_6H_3$	60	93	>300 (>300[33])
8i	4-ClC_6H_4	40	93	297-299 (298-300 [37])
8j	2,4-$(CH_3O)_2C_6H_3$	20	87	
8k	3,4-$OCH_2OC_6H_3$	30	86	>300 (>300[35])
8l	Pyrid-3-yl	80	76	
8m	Thiophen-2-yl	90	92	
8n	isobutyl	70	70	
8o	isopropyl	90	60	

The formation of **4**, **6** and **8** were characterized by spectroscopic analysis. Thus, the IR spectra of compounds **4**, **6** and **8** measured in potassium bromide pellets show one band of the elongation vibrations of the C=O groups at about 1640 cm^{-1} and one band of NH groups at about 3200 cm^{-1} for compounds **8**. In the ^1H NMR spectra of compounds **4**, **6** and **8** measured in dimethyl-d_6 sulfoxide were observed two signals of CH_3 groups at about 0.7-1.0 ppm, the signal of CH group at about 4.9 ppm. The structures of **4b** and **8h** were further confirmed by single crystal X-ray diffraction analysis. Figure 1 and Figure 2 show the molecular structures of **4b** and **8h**, respectively. The crystallographic data of these compounds are summarized in Table 4.

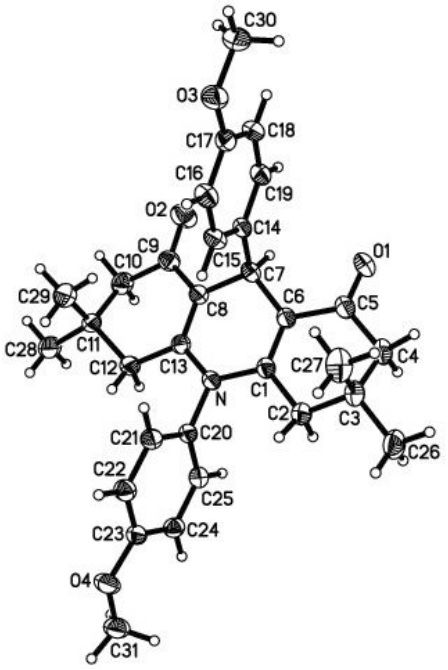


Figure 1. X-ray structure of **4b**.

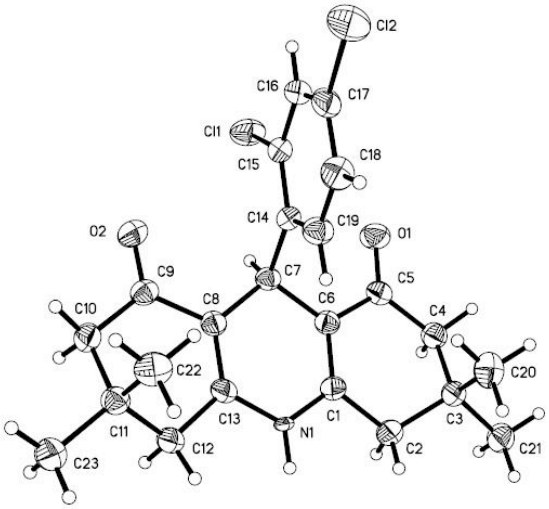
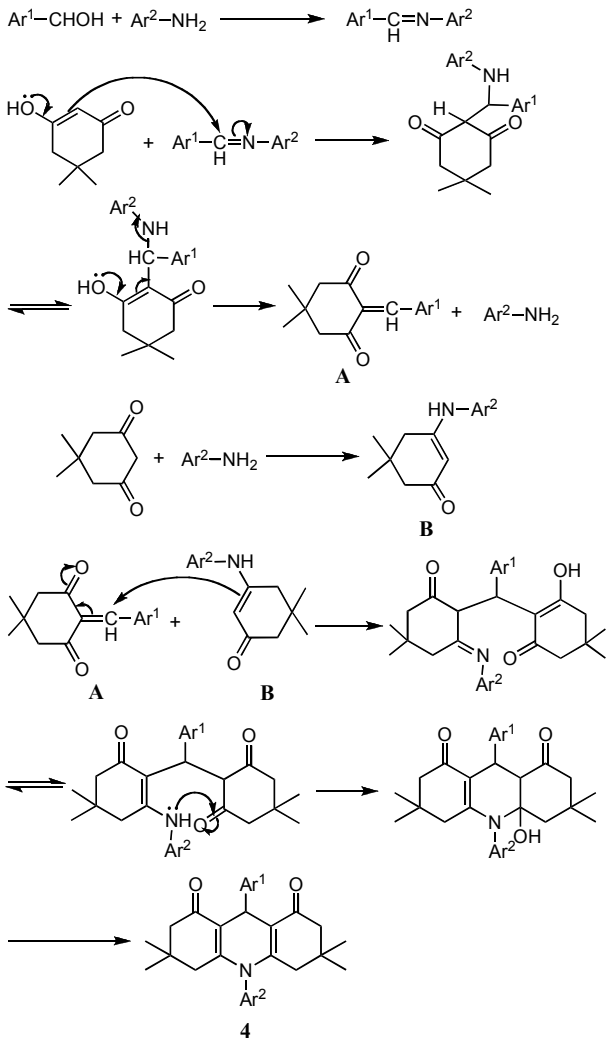


Figure 2. X-ray structure of **8h**.

Though the detailed mechanism of these reactions has not been clarified yet, the formation of **4** can be explained by the possible mechanism presented in Scheme 4. The reaction occurs *via* an initial formation of the imine from the condensation of aldehyde and amine, which suffers nucleophilic attack by 5,5-dimethyl-1,3-cyclohexanedione and loses amine to give the intermediate [**A**]. Condensation of another 5,5-dimethyl-1,3-cyclo- hexanedione and amine are taken place and to give another intermediate enamines [**B**]. Then, the Michael additation, cyclization and dehydration between intermediates [**A**] and [**B**] are taken place and to give products **4**.

Scheme 4



Evidence supporting this proposed mechanism came from the observation that when **9a** and **2** were treated under same reaction conditions, the expected product **4a** was obtained in a yield similar to that obtained in the one-pot reaction (Scheme 5).

Scheme 5

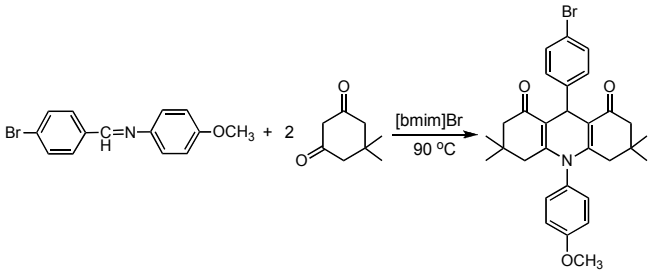


Table 4

Crystallographic Data for **4b** and **8h**.

	4b	**8h**
Empirical formula	$C_{31}H_{35}NO_4$	$C_{23}H_{25}Cl_2NO_2$
Formula weight	485.60	418.34
Temperature (K)	296(2)	298(2)
Wavelength (Å)	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic
Space group	$P2_1/c$	$P2_1/c$
a (Å)	12.089(2)	9.826(3)
b (Å)	11.447(2)	19.866(5)
c (Å)	19.742(3)	11.471(3)
α (°)	90	90
β (°)	101.00(1)	111.929(4)
γ (°)	90	90
V (Å^3)	2681.74(81)	2077.3(10)
Z	4	4
$D_{calc.}$ (Mg/m^3)	1.203	1.338
Absorption coefficient (mm^{-1})	0.079	0.331
F (000)	1040	880
Crystal size (mm)	0.58 × 0.38 ×0.30	0.34 × 0.29 ×0.16
θ Range (°)	1.72 to 25.50	2.05 to 25.01
Limiting indices	$0 \le h \le 14$	$-10 \le h \le 11$
	$0 \le k \le 13$	$-23 \le k \le 23$
	$-23 \le l \le 23$	$-13 \le l \le 8$
Reflections collected	5706	10831
Independent reflections	4988	3666
Data / restraints / parameters	4988 / 0 / 332	3666 / 0 / 253
Goodness-of-fit on F^2	0.908	1.012
Final *R* indices [$I > 2\sigma(I)$]	$R_1 = 0.0383$	$R_1 = 0.0551$
	$wR = 0.0854$	$wR = 0.1264$
R indices (all data)	$R_1 = 0.0693$	$R_1 = 0.1201$
	$wR = 0.0923$	$wR = 0.1631$
Largest diff. Peak and hole (e·Å^{-3})	0.143 and -0.134	0.407 and -0.493

In conclusion, a series of 3,3,6,6-tetramethyl-9,10-diaryl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-diones and 3,3,6,6-tetramethyl-9-aryl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-diones were synthesized by three-component reaction of aldehydes, 5,5-dimethyl-1,3-cyclohexanedione and aromatic amines or ammonium acetate in [bmim]Br. The advantages of this method are easier work-up, milder reaction conditions, high yields and environmentally benign procedure.

EXPERIMENTAL

Melting points were determined in open capillaries and are uncorrected. IR spectra were recorded on a Tensor 27 spectrometer in KBr. ^{1}H NMR and ^{13}C NMR spectra were measured on a Bruker DPX-400 MHz spectrometer using TMS as internal standard and DMSO-d_6 as solvent. Microanalyses were carried out on a Perkin-Elmer 2400 II instrument. X-ray diffraction was recorded on a Siemens P4 diffractometer.

General Procedure for the Three-component Reaction of Aldehydes 1, 5,5-Dimethyl-1,3-cyclohexanedione (2), and Aromatic Amines 3. A dry 50 mL flask was charged with aromatic aldehyde **1** (1 mmol), 5,5-dimethyl-1,3-cyclohexanedione **2** (2 mmol), aromatic amine **3** (1 mmol) and [bmim]Br (10 mL). The mixture was stirred at 90 °C for 2-4.5 h to complete the reaction (monitored by TLC), then cooled to room temperature. The yellow solid was collected by filtration and washed with water. The filtrate of [bmim]Br was then recovered for reuse by drying at 80 °C several hours in a vacuum. The crude product was purified by recrystallization from 95 % EtOH to give **4**.

3,3,6,6-Tetramethyl-9-(4-bromophenyl)-10-(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4a). This compound was obtained as solid with mp 247-248 °C; ir (potassium bromide): 2959, 1638, 1572, 1511, 1484, 1360, 1295, 1250, 1221, 1174, 1143, 1121, 1067, 1032, 1010, 846 cm^{-1}; ^{1}H nmr (DMSO-d_6): δ 0.72 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.80 (d, *J* = 17.6 Hz, 2H, 2 × CH), 2.00 (d, *J* = 16.0 Hz, 2H, 2 × CH), 2.19 (d, *J* = 16.0 Hz, 2H, 2 × CH), 2.20 (d, *J* = 17.6 Hz, 2H, 2 × CH), 3.86 (s, 3H, CH_3O), 5.00 (s, 1H, CH), 7.13 (d, *J* = 8.8 Hz, 2H, ArH), 7.25 (d, *J* = 8.4 Hz, 2H, ArH), 7.28-7.39 (m, 2H, ArH), 7.44 (d, *J* = 8.4 Hz, 2H, ArH). ^{13}C nmr (DMSO-d_6): δ 26.88, 29.95, 32.49, 32.61, 50.22, 56.16, 113.18, 115.67, 115.82, 119.42, 130.50, 131.46, 131.54, 146.38, 151.77, 160.01, 195.73. *Anal.* Calcd. for $C_{30}H_{32}BrNO_3$: C, 67.41; H, 6.03; N, 2.62. Found: C, 67.59; H, 5.97; N, 2.71.

3,3,6,6-Tetramethyl-9,10-bis(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4b). This compound was obtained as solid with mp 210-211 °C; ir (potassium bromide): 2955, 2838, 1647, 1607, 1575, 1508, 1458, 1440, 1421, 1362, 1296, 1221, 1174, 1140, 1120, 1030, 1000, 850, 837 cm^{-1}; ^{1}H nmr (DMSO-d_6): δ 0.73 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.79 (d, *J* = 17.2 Hz, 2H, 2 × CH), 1.99 (d, *J* = 16.0 Hz, 2H, 2 × CH), 2.18 (d, *J* = 16.0 Hz, 2H, 2 × CH), 2.19 (d, *J* = 17.2 Hz, 2H, 2 × CH), 3.69 (s, 3H, CH_3O), 3.86 (s, 3H, CH_3O), 4.98 (s, 1H, CH), 6.80 (d, *J* = 8.8 Hz, 2H, ArH), 7.12 (d, *J* = 8.8 Hz, 2H, ArH), 7.20 (d, *J* = 8.8 Hz, 2H, ArH), 7.36 (s, 2H, ArH). *Anal.* Calcd. for $C_{31}H_{35}NO_4$: C, 76.67; H, 7.26; N, 2.88. Found: C, 76.78; H, 7.15; N, 2.94.

3,3,6,6-Tetramethyl-9-(3,4-methylenedioxylphenyl)-10-(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4c). This compound was obtained as solid with mp 227-229 °C; ir (potassium bromide): 2954, 2903, 2842, 1639, 1574, 1510, 1478, 1440, 1360, 1295, 1250, 1222, 1140, 1121, 1087, 999, 943, 921, 867, 850, 815, 805, 780 cm^{-1}; ^{1}H nmr (DMSO-d_6): δ 0.75 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.81 (d, *J* = 17.2 Hz, 2H, 2 × CH), 2.02 (d, *J* = 16.0 Hz, 2H, 2 × CH), 2.18 (d, *J* = 16.0 Hz, 2H, 2 × CH), 2.19 (d, *J* = 17.2 Hz, 2H, 2 × CH), 3.85 (s, 3H, CH_3O), 4.96 (s, 1H, CH), 5.94 (s, 2H, OCH_2O), 6.74-6.79 (m, 3H, ArH), 7.13 (d, *J* = 8.8 Hz, 2H, ArH), 7.18-7.38 (m, 2H, ArH). *Anal.* Calcd. for $C_{31}H_{33}NO_5$: C, 74.53; H, 6.66; N, 2.80. Found: C, 74.48; H, 6.69; N, 2.86.

3,3,6,6-Tetramethyl-9-(4-methylphenyl)-10-(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4d). This compound was obtained as solid with mp 247-248 °C; ir (potassium bromide): 2959, 2841, 1640, 1575, 1511, 1466, 1424, 1360, 1310, 1294, 1277, 1245, 1213, 1175, 1141, 1120, 1029, 1000, 849, 835 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.72 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.79 (d, J = 17.6 Hz, 2H, 2 × CH), 1.99 (d, J = 16.0 Hz, 2H, 2 × CH), 2.18 (d, J = 16.0 Hz, 2H, 2 × CH), 2.20 (d, J = 17.6 Hz, 2H, 2 × CH), 2.22 (s, 3H, CH_3), 3.86 (s, 3H, CH_3O), 5.00 (s, 1H, CH), 7.04 (d, J = 7.6 Hz, 2H, ArH), 7.13 (d, J = 8.0 Hz, 2H, ArH), 7.18 (d, J = 8.0 Hz, 2H, ArH), 7.20-7.40 (m, 2H, ArH). *Anal.* Calcd. for $C_{31}H_{35}NO_3$: C, 79.28; H, 7.51; N, 2.98. Found: C, 79.35; H, 7.43; N, 3.06.

3,3,6,6-Tetramethyl-9-(3,4-dimethoxyphenyl)-10-(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4e). This compound was obtained as solid with mp 247-248 °C; ir (potassium bromide): 2955, 2837, 1636, 1573, 1510, 1465, 1419, 1363, 1311, 1294, 1248, 1213, 1174, 1138, 1024, 977, 928, 853, 815, 775, 754 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.75 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.81 (d, J = 17.2 Hz, 2H, 2 × CH), 2.02 (d, J = 16.0 Hz, 2H, 2 × CH), 2.19 (d, J = 16.0 Hz, 2H, 2 × CH), 2.21 (d, J = 17.2 Hz, 2H, 2 × CH), 3.69 (s, 3H, CH_3O), 3.71 (s, 3H, CH_3O), 3.85 (s, 3H, CH_3O), 5.00 (s, 1H, CH), 6.79-6.84 (m, 3H, ArH), 7.14 (s, 3H, ArH), 7.37 (s, 1H, ArH). *Anal.* Calcd. for $C_{32}H_{37}NO_5$: C, 74.54; H, 7.23; N, 2.72. Found: C, 74.63; H, 7.19; N, 2.64.

3,3,6,6-Tetramethyl-9-(4-(dimethylamino)phenyl)-10-(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4f). This compound was obtained as solid with mp 274-276 °C; ir (potassium bromide): 2961, 1652, 1603, 1577, 1518, 1461, 1370, 1295, 1250, 1223, 1169, 1132, 1113, 1026, 999, 979, 945, 887, 850, 827, 778, 745 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.74 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.79 (d, J = 17.2 Hz, 2H, 2 × CH), 1.99 (d, J = 16.0 Hz, 2H, 2 × CH), 2.17 (d, J = 16.0 Hz, 2H, 2 × CH), 2.19 (d, J = 17.2 Hz, 2H, 2 × CH), 3.22 (s, 6H, $(CH_3)_2N$), 3.86 (s, 3H, CH_3O), 4.92 (s, 1H, CH), 6.61 (d, J = 8.4 Hz, 2H, ArH), 7.10-7.14 (m, 4H, ArH), 7.20-7.36 (m, 2H, ArH). *Anal.* Calcd. for $C_{32}H_{38}N_2O_3$: C, 77.08; H, 7.68; N, 5.62. Found: C, 77.23; H, 7.62; N, 5.58.

3,3,6,6-Tetramethyl-9-(4-chlorophenyl)-10-(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4g). This compound was obtained as solid with mp 274-276 °C; ir (potassium bromide): 2956, 1646, 1572, 1510, 1471, 1360, 1295, 1242, 1222, 1169, 1143, 1120, 1036, 1003, 934, 886, 846, 798, 750, 718, 700, 662 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.74 (s, 6H, 2 × CH_3), 0.87 (s, 6H, 2 × CH_3), 1.79 (d, J = 17.2 Hz, 2H, 2 × CH), 1.94 (d, J = 16.4 Hz, 2H, 2 × CH), 2.15 (d, J = 16.4 Hz, 4H, 4 × CH), 3.86 (s, 3H, CH_3O), 5.27 (s, 1H, CH), 7.08-7.17 (m, 3H, ArH), 7.22-7.38 (m, 4H, ArH), 7.50 (d, J = 8.0 Hz, 1H, ArH). *Anal.* Calcd. for $C_{30}H_{32}ClNO_3$: C, 73.53; H, 6.58; N, 2.86. Found: C, 73.66; H, 6.51; N, 2.82.

3,3,6,6-Tetramethyl-9-(3,4-dichlorophenyl)-10-(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4h). This compound was obtained as solid with mp 209-211 °C; ir (potassium bromide): 2959, 1645, 1573, 1513, 1456, 1376, 1294, 1250, 1206, 1169, 1132, 1027, 932, 876, 847, 779, 745, 702, 673 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.73 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.83 (d, J = 17.2 Hz, 2H, 2 × CH), 2.03 (d, J = 16.0 Hz, 2H, 2 × CH), 2.20 (d, J = 16.0 Hz, 2H, 2 × CH), 2.21 (d, J = 17.2 Hz, 2H, 2 × CH), 3.86 (s, 3H, CH_3O), 5.00 (s, 1H, CH), 7.14 (d, J = 7.2 Hz, 2H, ArH), 7.24 (s, 1H, ArH), 7.27 (dd, J_1 = 2.0 Hz, J_2 = 8.4 Hz, 1H, ArH), 7.37 (s, 1H, ArH), 7.45 (d, J = 2.0 Hz, 1H, ArH), 7.54 (d, J = 8.4 Hz, 1H, ArH). *Anal.* Calcd. for $C_{30}H_{31}Cl_2NO_3$: C, 68.70; H, 5.96; N, 2.67. Found: C, 68.82; H, 6.04; N, 2.75.

3,3,6,6-Tetramethyl-9-(4-hydroxyphenyl)-10-(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4i). This compound was obtained as solid with mp > 300 °C; ir (potassium bromide): 3234, 2960, 2873, 1634, 1593, 1567, 1510, 1452, 1366, 1313, 1295, 1251, 1213, 1168, 1142, 1121, 1107, 1030, 1001, 887, 844, 745 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.72 (s, 6H, 2 × CH_3), 0.88 (s, 6H, 2 × CH_3), 1.78 (d, J = 17.2 Hz, 2H, 2 × CH), 1.99 (d, J = 16.0 Hz, 2H, 2 × CH), 2.17 (d, J = 16.0 Hz, 2H, 2 × CH), 2.18 (d, J = 17.2 Hz, 2H, 2 × CH), 3.85 (s, 3H, CH_3O), 4.93 (s, 1H, CH), 6.61 (d, J = 8.4 Hz, 2H, ArH), 7.07 (d, J = 8.4 Hz, 2H, ArH), 7.12 (d, J = 8.4 Hz, 2H, ArH), 7.21 (s, 1H, ArH), 7.36 (s, 1H, ArH), 9.09 (s, 1H, OH). *Anal.* Calcd. for $C_{30}H_{33}NO_4$: C, 76.41; H, 7.05; N, 2.97. Found: C, 76.59; H, 6.97; N, 3.06.

3,3,6,6-Tetramethyl-9-(4-bromophenyl)-10-(3-chloro-4-methylphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4j). This compound was obtained as solid with mp 293-295 °C; ir (potassium bromide): 2957, 1652, 1603, 1567, 1495, 1372, 1301, 1264, 1224, 1176, 1143, 1117, 1068, 1055, 1009, 923, 887, 838, 775, 731, 708 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.73 (s, 6H, 2 × CH_3), 0.90 (s, 6H, 2 × CH_3), 1.80 (d, J = 16.0 Hz, 2H, 2 × CH), 2.01 (d, J = 16.0 Hz, 2H, 2 × CH), 2.18 (d, J = 16.0 Hz, 4H, 4 × CH), 2.44 (s, 3H, CH_3), 4.99 (s, 1H, CH), 7.27 (d, J = 8.4 Hz, 2H, ArH), 7.34-7.37 (m, 1H, ArH), 7.43 (d, J = 8.4 Hz, 2H, ArH), 7.59 (d, J = 8.0 Hz, 2H, ArH). ^{13}C nmr (DMSO-d_6): δ 19.40, 26.17, 29.19, 31.96, 40.87, 49.51, 112.61, 125.79, 129.77, 129.85, 130.57, 130.69, 136.95, 137.01, 145.34, 150.12, 150.83, 151.19, 194.74. *Anal.* Calcd. for $C_{30}H_{31}BrClNO_2$: C, 65.17; H, 5.65; N, 2.53. Found: C, 65.28; H, 5.60; N, 2.57.

3,3,6,6-Tetramethyl-9-(4-chlorophenyl)-10-(3-chloro-4-methylphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4k). This compound was obtained as solid with mp 288-290 °C; ir (potassium bromide): 2958, 2870, 1642, 1601, 1578, 1489, 1472, 1361, 1301, 1261, 1221, 1176, 1143, 1122, 1089, 1054, 1014, 1000, 924, 887, 840 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.72 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.80 (d, J = 16.0 Hz, 2H, 2 × CH), 2.00 (d, J = 16.0 Hz, 2H, 2 × CH), 2.18 (d, J = 16.0 Hz, 4H, 4 × CH), 2.44 (s, 3H, CH_3), 5.00 (s, 1H, CH), 7.27-7.34 (m, 5H, ArH), 7.59 (d, J = 8.0 Hz, 2H, ArH). *Anal.* Calcd. for $C_{30}H_{31}Cl_2NO_2$: C, 70.86; H, 6.15; N, 2.75. Found: C, 70.92; H, 6.11; N, 2.84.

3,3,6,6-Tetramethyl-9-(4-methylphenyl)-10-(3-chloro-4-methylphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4l). This compound was obtained as solid with mp 248-250 °C; ir (potassium bromide): 2957, 2869, 1642, 1602, 1578, 1495, 1472, 1361, 1301, 1260, 1221, 1177, 1143, 1122, 1053, 999, 886, 835, 715, 702 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.73 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.79 (d, J = 16.0 Hz, 2H, 2 × CH), 2.00 (d, J = 16.0 Hz, 2H, 2 × CH), 2.17 (d, J = 16.0 Hz, 4H, 4 × CH), 2.22 (s, 3H, CH_3), 2.44 (s, 3H, CH_3), 4.99 (s, 1H, CH), 7.02-7.05 (m, 3H, ArH), 7.19 (d, J = 8.0 Hz, 2H, ArH), 7.31 (s, 1H, ArH), 7.59 (d, J = 8.0 Hz, 1H, ArH). *Anal.* Calcd. for $C_{31}H_{34}ClNO_2$: C, 76.29; H, 7.02; N, 2.87. Found: C, 76.40; H, 6.98; N, 2.94.

3,3,6,6-Tetramethyl-9-(thiophen-2-yl)-10-(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4m). This compound was obtained as solid with mp 240-242 °C; ir (potassium bromide): 3064, 2959, 2837, 1651, 1573, 1506, 1441, 1371, 1293, 1264, 1213, 1173, 1121, 1074, 1030, 997, 980, 921, 888, 843, 820, 772, 744, 699 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.78 (s, 6H, 2 × CH_3), 0.91 (s, 6H, 2 × CH_3), 1.78 (d, J = 17.6 Hz, 2H, 2 × CH), 2.07 (d, J = 16.0 Hz, 2H, 2 × CH), 2.23

(d, J = 16.0 Hz, 2H, 2 × CH), 2.24 (d, J = 17.6 Hz, 2H, 2 × CH), 3.85 (s, 3H, CH_3O), 5.37 (s, 1H, CH), 6.79 (d, J = 3.2 Hz, 1H, ArH), 6.87 (dd, J_1 = 3.2 Hz, J_2 = 5.6 Hz, 1H, ArH), 7.12 (s, 3H, ArH), 7.22 (d, J = 5.6 Hz, 1H, ArH), 7.37-7.43 (m, 1H, ArH). *Anal*. Calcd. for $C_{28}H_{31}NO_3S$: C, 72.85; H, 6.77; N, 3.03. Found: C, 72.82; H, 6.85; N, 3.06.

3,3,6,6-Tetramethyl-9-(pyridine-3-yl)-10-(4-methoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4n). This compound was obtained as solid with mp 231-232 °C; ir (potassium bromide): 2956, 1641, 1575, 1508, 1473, 1426, 1362, 1294, 1248, 1222, 1170, 1144, 1123, 1108, 1027, 1003, 853, 716 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.71 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.83 (d, J = 17.6 Hz, 2H, 2 × CH), 2.01 (d, J = 16.0 Hz, 2H, 2 × CH), 2.20 (d, J = 16.0 Hz, 4H, 4 × CH), 3.86 (s, 3H, CH_3O), 5.01 (s, 1H, CH), 7.13 (d, J = 7.2 Hz, 2H, ArH), 7.28-7.37 (m, 3H, ArH), 7.65-7.67 (m, 1H, ArH), 8.31 (s, 1H, ArH), 8.52 (s, 1H, ArH). *Anal*. Calcd. for $C_{29}H_{32}N_2O_3$: C, 76.29; H, 7.06; N, 6.14. Found: C, 76.37; H, 7.01; N, 6.09.

3,3,6,6-Tetramethyl-9-(4-chlorophenyl)-10-phenyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4o). This compound was obtained as solid with mp 231-233 °C; ir (potassium bromide): 3060, 2957, 2931, 2869, 1640, 1592, 1576, 1490, 1472, 1452, 1361, 1300, 1276, 1262, 1221, 1176, 1143, 1120, 1088, 1012, 980, 939, 888, 940, 771, 704 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.71 (s, 6H, 2 × CH_3), 0.82 (s, 6H, 2 × CH_3), 1.76 (d, J = 17.6 Hz, 2H, 2 × CH), 2.01 (d, J = 16.0 Hz, 2H, 2 × CH), 2.20 (d, J = 16.0 Hz, 2H, 2 × CH), 2.21 (d, J = 17.6 Hz, 2H, 2 × CH), 5.03 (s, 1H, CH), 7.29-7.35 (m, 4H, ArH), 7.38-7.48 (m, 2H, ArH), 7.55-7.66 (m, 3H, ArH). ^{13}C nmr (DMSO-d_6): δ 26.82, 29.91, 32.43, 32.64, 50.21, 113.26, 128.43, 128.55, 130.08, 130.13, 130.78, 130.95, 139.03, 145.80, 151.19, 195.71. *Anal*. Calcd. for $C_{29}H_{30}ClNO_2$: C, 75.72; H, 6.57; N, 3.04. Found: C, 75.82; H, 6.64; N, 2.98.

3,3,6,6-Tetramethyl-9-(4-chlorophenyl)-10-(4-fluorophenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4p). This compound was obtained as solid with mp 296-298 °C; ir (potassium bromide): 2955, 2933, 2869, 1639, 1577, 1541, 1507, 1486, 1472, 1417, 1361, 1302, 1279, 1262, 1220, 1177, 1144, 1118, 1086, 1012, 980, 888, 861, 840, 805, 743 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.72 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.77 (d, J = 17.6 Hz, 2H, 2 × CH), 2.01 (d, J = 16.0 Hz, 2H, 2 × CH), 2.19 (d, J = 16.0 Hz, 2H, 2 × CH), 2.20 (d, J = 17.6 Hz, 2H, 2 × CH), 5.02 (s, 1H, CH), 7.28-7.34 (m, 4H, ArH), 7.43-7.59 (m, 4H, ArH). ^{13}C nmr (DMSO-d_6): δ 26.82, 29.91, 32.41, 32.64, 50.21, 113.37, 128.54, 130.11, 133.10, 135.14, 136.28, 135.32, 135.39, 145.81, 151.26, 195.73. *Anal*. Calcd. for $C_{29}H_{29}ClFNO_2$: C, 72.87; H, 6.12; N, 2.93. Found: C, 72.94; H, 6.08; N, 2.97.

3,3,6,6-Tetramethyl-9-(4-methoxyphenyl)-10-(4-chlorophenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4q). This compound was obtained as solid with mp 269-271 °C; ir (potassium bromide): 3052, 2953, 2837, 1641, 1578, 1508, 1491, 1473, 1439, 1361, 1299, 1259, 1220, 1173, 1141, 1120, 1105, 1089, 1037, 1016, 999, 886, 864, 850, 834, 816, 780, 740, 720 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.73 (s, 6H, 2 × CH_3), 0.89 (s, 6H, 2 × CH_3), 1.76 (d, J = 17.2 Hz, 2H, 2 × CH), 2.00 (d, J = 16.0 Hz, 2H, 2 × CH), 2.18 (d, J = 16.0 Hz, 2H, 2 × CH), 2.19 (d, J = 17.2 Hz, 2H, 2 × CH), 3.69 (s, 3H, CH_3O), 4.97 (s, 1H, CH), 6.80 (d, J = 8.8 Hz, 2H, ArH), 7.12 (d, J = 8.8 Hz, 2H, ArH), 7.38-7.53 (m, 2H, ArH), 7.69 (d, J = 8.8 Hz, 2H, ArH). *Anal*. Calcd. for $C_{30}H_{32}ClNO_3$: C, 73.53; H, 6.58; N, 2.86. Found: C, 73.65; H, 6.63; N, 2.89.

3,3,6,6-Tetramethyl-9-(4-methylphenyl)-10-(2-methylphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (4r). This compound was obtained as solid with mp 242-245 °C; ir (potassium bromide): 3050, 2950, 1638, 1577, 1510, 1492, 1472, 1423, 1360, 1298, 1258, 1222, 1176, 1140, 1120, 1020, 998, 937, 920, 835, 767, 739, 717, 668 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.75 (s, 6H, 2 × CH_3), 0.87 (s, 6H, 2 × CH_3), 1.50 (d, J = 17.2 Hz, 2H, 2 × CH), 1.99 (d, J = 16.4 Hz, 2H, 2 × CH), 2.16-2.26 (m, 10H, 2 × CH_3, 4 × CH), 5.03 (s, 1H, CH), 7.02 (d, J = 8.0 Hz, 2H, ArH), 7.21 (d, J = 8.0 Hz, 2H, ArH), 7.34 (d, J = 8.0 Hz, 1H, ArH), 7.39-7.46 (m, 1H, ArH), 7.47-7.52 (m, 2H, ArH). ^{13}C nmr (DMSO-d_6): δ 18.41, 21.22, 26.37, 30.33, 32.37, 41.84, 50.24, 113.59, 128.16, 128.44, 128.90, 130.32, 130.81, 131.98, 135.18, 137.00, 138.10, 144.24, 150.50, 195.66. *Anal*. Calcd. for $C_{31}H_{35}NO_2$: C, 82.08; H, 7.78; N, 3.09. Found: C, 82.21; H, 7.74; N, 3.15.

General Procedure for the Three-component Reaction of Dicarboxyldehydes 5, 5,5-Dimethyl-1,3-cyclohexanedione (2), and Aromatic amines 3. A dry 50 mL flask was charged with dicarboxaldehyde **5** (1 mmol), 5,5-dimethyl-1,3-cyclohexanedione **2** (4 mmol), aromatic amine **3** (2 mmol) and [bmim]Br (10 mL). The mixture was stirred at 90 °C for 2.5-3 h to complete the reaction (monitored by TLC), then cooled to room temperature. The yellow solid was filtered off and washed with water. The crude product was purified by recrystallization from 95 % EtOH to give **6**.

9,9'-(1,3-Phenylene)bis(10-(4-methoxyphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione) (6a). This compound was obtained as solid with mp 278-280 °C; ir (potassium bromide): 2956, 2870, 1638, 1574, 1511, 1471, 1364, 1294, 1250, 1222, 1174, 1142, 1121, 1027, 846 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.70 (s, 12H, 4 × CH_3), 0.88 (s, 12H, 4 × CH_3), 1.81 (d, J = 17.6 Hz, 4H, 4 × CH), 1.93 (d, J = 16.0 Hz, 4H, 4 × CH), 2.17 (d, J = 16.0 Hz, 4H, 4 × CH), 2.21 (d, J = 17.6 Hz, 4H, 4 × CH), 3.86 (s, 6H, 2 × CH_3O), 5.02 (s, 2H, 2 × CH), 7.01-7.14 (m, 7H, ArH), 7.32-7.45 (m, 3H, ArH), 7.58-7.66 (m, 2H, ArH). ^{13}C nmr (DMSO-d_6): δ 26.85, 30.27, 32.49, 41.75, 50.40, 56.12, 113.62, 115.49, 115.59, 115.64, 125.37, 128.16, 131.88, 146.54, 151.28, 159.90, 195.47. *Anal*. Calcd. for $C_{54}H_{60}N_2O_6$: C, 77.85; H, 7.26; N, 3.36. Found: C, 77.97; H, 7.15; N, 3.42.

9,9'-(1,3-Phenylene)bis(10-(4-methylphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione) (6b). This compound was obtained as solid with mp 261-263 °C; ir (potassium bromide): 2954, 2869, 1639, 1576, 1512, 1469, 1365, 1300, 1260, 1221, 1176, 1142, 1121, 1019, 999, 919, 889, 845, 814, 734, 703 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.70 (s, 12H, 4 × CH_3), 0.88 (s, 12H, 4 × CH_3), 1.79 (d, J = 17.2 Hz, 4H, 4 × CH), 1.93 (d, J = 16.0 Hz, 4H, 4 × CH), 2.17 (d, J = 16.0 Hz, 4H, 4 × CH), 2.21 (d, J = 17.2 Hz, 4H, 4 × CH), 2.43 (s, 6H, 2 × CH_3), 5.03 (s, 2H, 2 × CH), 7.01-7.12 (m, 3H, ArH), 7.35-7.44 (m, 7H, ArH), 7.49-7.60 (m, 2H, ArH). *Anal*. Calcd. for $C_{54}H_{60}N_2O_4$: C, 80.96; H, 7.55; N, 3.50. Found: C, 81.04; H, 7.47; N, 3.58.

9,9'-(1,3-Phenylene)bis(10-phenyl-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione) (6c). This compound was obtained as solid with mp > 300 °C; ir (potassium bromide): 3059, 2956, 2870, 1657, 1592, 1491, 1452, 1368, 1297, 1260, 1220, 1176, 1142, 1120, 1081, 1021, 1000, 979, 888, 813 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.70 (s, 12H, 4 × CH_3), 0.87 (s, 12H, 4 × CH_3), 1.76 (d, J = 17.6 Hz, 4H, 4 × CH), 1.94 (d, J = 16.0 Hz, 4H, 4 × CH), 2.18 (d, J = 16.0 Hz, 4H, 4 × CH), 2.22 (d, J = 17.6 Hz, 4H, 4 × CH), 5.04 (s, 2H, 2 ×

CH), 7.03-7.11 (m, 3H, ArH), 7.46-7.65 (m, 11H, ArH). *Anal.* Calcd. for $C_{52}H_{56}N_2O_4$: C, 80.80; H, 7.30; N, 3.62. Found: C, 80.96; H, 7.20; N, 3.69.

9,9'-(1,4-Phenylene)bis(10-(4-methoxyphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione) (6d). This compound was obtained as solid with mp > 300 °C; ir (potassium bromide): 2956, 2870, 1637, 1576, 1510, 1466, 1419, 1363, 1293, 1249, 1213, 1175, 1142, 1120, 1022, 979, 888, 846, 824 cm^{-1}; ^1H nmr (DMSO-d_6): 0.66 (s, 12H, 4 × CH_3), 0.87 (s, 12H, 4 × CH_3), 1.79 (d, J = 17.6 Hz, 4H, 4 × CH), 1.99 (d, J = 16.0 Hz, 4H, 4 × CH), 2.16 (d, J = 16.0 Hz, 4H, 4 × CH), 2.18 (d, J = 17.6 Hz, 4H, 4 × CH), 3.86 (s, 6H, 2 × CH_3O), 5.01 (s, 2H, 2 × CH), 7.12-7.41 (m, 12H, ArH). *Anal.* Calcd. for $C_{54}H_{60}N_2O_6$: C, 77.85; H, 7.26; N, 3.36. Found: C, 78.02; H, 7.19; N, 3.48.

General Procedure for the Three-component Reaction of Aldehydes 1, 5,5-Dimethyl-1,3-cyclohexanedione (2), and Ammonium Acetate (2). A dry 50 mL flask was charged with aldehyde **1** (1 mmol), 5,5-dimethyl-1,3-cyclohexanedione (2 mmol), ammonium acetate (10 mmol) and [bmim]Br (10 mL). The mixture was stirred at 90 °C for 15-90 min to complete the reaction (monitored by TLC), then cooled to room temperature. The yellow solid was filtered off and washed with water. The crude product was purified by recrystallization from 95 % EtOH to give **8**.

3,3,6,6-Tetramethyl-9-(3,4-dichlorophenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (8g). This compound was obtained as solid with mp > 300 °C; ir (potassium bromide): 3177, 3061, 2957, 2810, 1647, 1611, 1491, 1396, 1362, 1259, 1221, 1142, 1029, 1009, 977, 880, 830, 762, 733, 701 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.87 (s, 6H, 2 × CH_3), 1.01 (s, 6H, 2 × CH_3), 2.01 (d, J = 16.0 Hz, 2H, 2 × CH), 2.19 (d, J = 16.0 Hz, 2H, 2 × CH), 2.35 (d, J = 16.8 Hz, 2H, 2 × CH), 2.47 (d, J = 16.8 Hz, 2H, 2 × CH), 4.78 (s, 1H, CH), 7.12 (dd, J_1 = 1.6 Hz, J_2 = 8.4 Hz, 1H, ArH), 7.31 (d, J = 1.6 Hz, 1H, ArH), 7.46 (d, J = 8.4 Hz, 1H, ArH), 9.43 (br. s, 1H, NH). ^{13}C nmr (DMSO-d_6): δ 27.16, 29.64, 32.83, 33.66, 50.80, 111.20, 128.60, 128.64, 130.29, 130.62, 130.73, 148.73, 150.51, 195.07. *Anal.* Calcd. for $C_{23}H_{25}Cl_2NO_2$: C, 66.03; H, 6.02; N, 3.35. Found: C, 66.21; H, 5.97; N, 3.42.

3,3,6,6-Tetramethyl-9-(2,4-dimethoxyphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (8j). This compound was obtained as solid with mp 267-269 °C; ir (potassium bromide): 3193, 3068, 2954, 2835, 1638, 1603, 1485, 1397, 1367, 1295, 1267, 1224, 1171, 1157, 1145, 1126, 1043, 930, 828 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.82 (s, 6H, 2 × CH_3), 1.00 (s, 6H, 2 × CH_3), 1.88 (d, J = 16.0 Hz, 2H, 2 × CH), 2.11 (d, J = 16.0 Hz, 2H, 2 × CH), 2.21 (d, J = 16.8 Hz, 2H, 2 × CH), 2.40 (d, J = 16.8 Hz, 2H, 2 × CH), 3.64 (s, 3H, CH_3O), 3.67 (s, 3H, CH_3O), 4.84 (s, 1H, CH), 6.31 (dd, J_1 = 2.4 Hz, J_2 = 8.4 Hz, 1H, ArH), 6.35 (d, J = 2.4 Hz, 1H, ArH), 7.05 (d, J = 8.4 Hz, 1H, ArH), 9.17 (br. s, 1H, NH). *Anal.* Calcd. for $C_{25}H_{31}NO_4$: C, 73.32; H, 7.63; N, 3.42. Found: C, 73.46; H, 7.58; N, 3.44.

3,3,6,6-Tetramethyl-9-(pyridine-3-yl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (8l). This compound was obtained as solid with mp > 300 °C; ir (potassium bromide): 3171, 3039, 2957, 1635, 1586, 1507, 1425, 1396, 1367, 1257, 1223, 1171, 1145, 1126, 1027, 1009, 833, 713 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.86 (s, 6H, 2 × CH_3), 1.02 (s, 6H, 2 × CH_3), 1.99 (d, J = 16.4 Hz, 2H, 2 × CH), 2.19 (d, J = 16.0 Hz, 2H, 2 × CH), 2.35 (d, J = 17.2 Hz, 2H, 2 × CH), 2.47 (d, J = 17.2 Hz, 2H, 2 × CH), 4.79 (s, 1H, CH), 7.21 (dd, J_1 = 4.8 Hz, J_2 = 7.6 Hz, 1H, ArH), 7.48 (dd, J_1 = 1.2 Hz, J_2 = 7.6 Hz, 1H, ArH), 8.24 (d, J = 4.4 Hz, 1H, ArH), 8.37 (s, 1H, ArH), 9.42 (s, 1H, NH). *Anal.* Calcd. for $C_{22}H_{26}N_2O_2$: C, 75.40; H, 7.48; N, 7.99. Found C, 75.62; H, 7.40; N, 8.05.

3,3,6,6-Tetramethyl-9-(thiophen-2-yl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (8m). This compound was obtained as solid with mp > 300 °C; ir (potassium bromide): 3278, 3211, 3065, 2956, 2931, 2872, 1638, 1625, 1602, 1482, 1395, 1371, 1250, 1218, 1168, 1140, 1122, 1031, 1003, 980, 887, 850, 716, 690 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.94 (s, 6H, 2 × CH_3), 1.03 (s, 6H, 2 × CH_3), 2.08 (d, J = 16.0 Hz, 2H, 2 × CH), 2.22 (d, J = 16.0 Hz, 2H, 2 × CH), 2.32 (d, J = 17.2 Hz, 2H, 2 × CH), 2.45 (d, J = 17.2 Hz, 2H, 2 × CH), 5.15 (s, 1H, CH), 6.65 (d, J = 3.2 Hz, 1H, ArH), 6.80 (dd, J_1 = 3.2 Hz, J_2 = 4.8 Hz, 1H, ArH), 7.14 (d, J = 4.8 Hz, 1H, ArH), 9.45 (s, 1H, NH). *Anal.* Calcd. for $C_{21}H_{25}NO_2S$: C, 70.95; H, 7.09; N, 3.94. Found C, 71.07; H, 7.15; N, 3.82.

3,3,6,6-Tetramethyl-9-(isobutyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (8n). This compound was obtained as solid with mp 254-256 °C; ir (potassium bromide): 3277, 3209, 3065, 2954, 2871, 1637, 1617, 1602, 1479, 1425, 1380, 1276, 1219, 1168, 1142, 1122, 1001, 978, 941, 886, 752, 729, 677 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.81 (d, J = 6.8 Hz, 6H, 2 × CH_3), 0.95 (t, J = 6.8 Hz, 2H, CH_2), 1.00 (s, 6H, 2 × CH_3), 1.02 (s, 6H, 2 × CH_3), 1.29-1.36 (m, 1H, CH), 2.07 (d, J = 16.0 Hz, 2H, 2 × CH), 2.17 (d, J = 16.4 Hz, 2H, 2 × CH), 2.25 (d, J = 17.2 Hz, 2H, 2 × CH), 2.38 (d, J = 17.2 Hz, 2H, 2 × CH), 3.82 (t, J = 6.8 Hz, 1H, CH), 9.18 (s, 1H, NH). ^{13}C nmr (DMSO-d_6): δ 23.07, 23.61, 23.84, 26.19, 29.27, 31.87, 46.31, 50.41, 111.78, 149.65, 194.32. Anal. Calcd. for $C_{21}H_{31}NO_2$: C, 76.55; H, 9.48; N, 4.25. Found C, 76.72; H, 9.43; N, 4.28.

3,3,6,6-Tetramethyl-9-(isopropyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (8o). This compound was obtained as solid with mp 283-284 °C; ir (potassium bromide): 3286, 3197, 3069, 2955, 2922, 2880, 1641, 1620, 1603, 1482, 1448, 1394, 1370, 1315, 1298, 1250, 1223, 1170, 1140, 1119, 1004, 975, 888, 745, 681 cm^{-1}; ^1H nmr (DMSO-d_6): δ 0.64 (d, J = 6.8 Hz, 6H, 2 × CH_3), 1.03 (s, 6H, 2 × CH_3), 1.05 (s, 6H, 2 × CH_3), 1.50-1.53 (m, 1H, CH), 2.11 (d, J = 16.0 Hz, 2H, 2 × CH), 2.18 (d, J = 16.4 Hz, 2H, 2 × CH), 2.27 (d, J = 17.2 Hz, 2H, 2 × CH), 2.39 (d, J = 17.2 Hz, 2H, 2 × CH), 3.80 (t, J = 4.0 Hz, 1H, CH), 9.12 (s, 1H, NH). ^{13}C nmr (DMSO-d_6): δ 19.84, 27.24, 30.12, 31.68, 32.42, 35.49, 51.21, 110.09, 151.56, 195.63. *Anal.* Calcd. for $C_{20}H_{29}NO_2$: C, 76.15; H, 9.27; N, 4.44. Found C, 76.33; H, 9.25; N, 4.39.

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