

A Green Procedure for the Synthesis of 1,8-Dioxodecahydroacridine Derivatives under Microwave Irradiation in Aqueous Media without Catalyst

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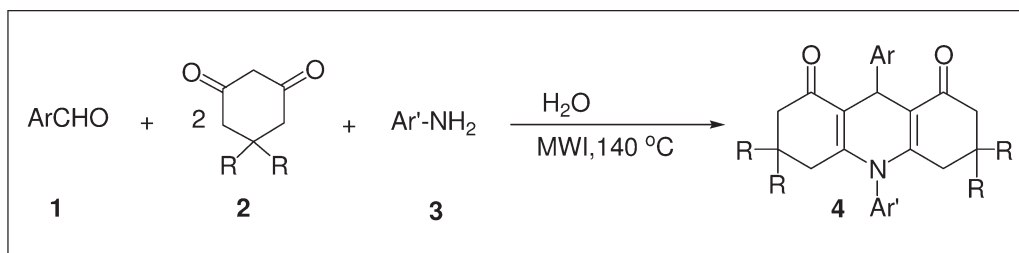
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A green procedure for the synthesis of 1,8-dioxo-decahydroacridine derivatives is developed under microwave irradiation without catalyst in water. This method provides several advantages such as excellent yields (86–96%), simple workup procedure, and environment friendliness.

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INTRODUCTION

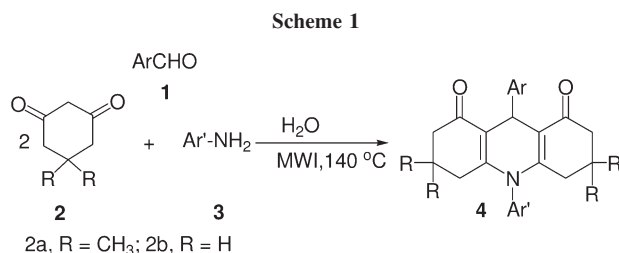
Water as a solvent has many advantages in organic synthesis, both from economic and from environmental points of view. Water has, therefore, become an attractive medium for many organic reactions. Many important types of heterocyclic compounds, such as triazines [1], acridines [2], quinolines [3], pyridines [4], indoles [5], pyrazines [6], furans [7], and pyrimidines [8] have been synthesized in aqueous media.

4-Aryl-1,4-dihydropyridines (1,4-DHPs) have proved to be valuable as drugs for the treatment of cardiovascular disorders [9] and constitute an important class of calcium channel blockers [10]. It is well established that slight structural modification on the DHP ring may result in remarkable change of pharmacological effect [11–14]. With a 1,4-DHP parent nucleus, acridine-1,8-diones have been shown to have very high lasing efficiencies [15] and used as photoinitiators [16]. Recently, many methods have become available for the synthesis of these important class of derivatives. We have synthesized these compounds from schiff's base and dimedone or 1,3-cyclohexanedione or three-component (aldehyde, dimedone, and arylamines) in glycol under microwave [17,18]. Jin et al. [19] reported that this reaction could

be carried out catalyzed by *p*-dodecylbenzenesulfonic acid (DBSA) in water. However, these reactions must be carried out by impetus of catalyst or in organic solvents. Wang et al. [20] reported the same reaction proceeded in an ionic medium, and Wang and Miao [21] achieved the reactions in aqueous solvent at traditional heating. The reaction time was 10 h, the yield was 72–75%. Arylamine used was only *p*-toluidine. Microwave-assisted organic reaction using water as solvent has peculiarity of “safe solvents” and “energy efficiency.” It was widely used in the organic synthesis [22]. In this article, we would like to report the green synthesis of 1,8-dioxo-9,10-diaryl-decahydroacridines without catalyst via the combination of aqueous solvent and microwave heating, using a variety of arylamines including the anilines contained electron-withdrawing groups and electron-donating groups (Scheme 1).

RESULTS AND DISCUSSION

When treating aldehyde **1** with dimedone or 1,3-cyclohexanedione **2** and primary arylamines **3** under microwave irradiation, the target compound **4** were obtained.



To demonstrate the efficiency and the applicability of this method, we investigated the reaction of a variety of aromatic aldehydes **1**, dimedone, and a variety of primary arylamines **3** at 140 °C in aqueous media. As shown in Table 1, a series of **1** and **3**, in which the aromatic ring contained electron-withdrawing groups (such as halo or nitro) or electron-donating groups (such as methyl or methyloxy), reacted with **2a** under the same reaction conditions to give the corresponding product **4** in good yields (entries 1–14). We thus concluded that there were no obvious electronic effect of the substituents on the aromatic rings. To further expand the scope of the present method, the replacement of dimedone **2a** with 1,3-cyclohexanedione **2b** was examined. To our delight, under the same conditions, the reactions proceeded steadily to afford a series of 1,8-dioxo-decahydroacridine derivatives in good yields (Table 1, entries 15–25).

The structures of all the synthesized compounds were established on the basis of their spectroscopic data (IR

and ¹H NMR). In addition, the X-ray diffraction analysis of product **4e** [23], **4k** [24], **4l** [25], **4n** [26], was carried out to confirm its structure. The crystal structure of **4k** is shown in Figure 1.

In conclusion, we have developed a green chemistry method for the synthesis of 1,8-dioxo-decahydroacridine derivatives. Excellent yields were obtained not only for anilines substituted with electron-donating groups but also for ones containing electron-withdrawing groups. The method avoided using organic solvents and had the main advantages of convenient procedure and environmental friendliness.

EXPERIMENTAL

Microwave irradiation was carried out with microwave oven Emrys™ Creator from Personal Chemistry, Uppsala, Sweden. Melting points were determined in open capillaries and were uncorrected. IR spectra were taken on a TENSOR 27 spectrometer in KBr and reported in cm⁻¹. ¹H NMR spectra were measured on a Bruke DPX 400 MHz spectrometer in DMSO-*d*₆ with chemical shift (δ) given in ppm relative to TMS as internal standard. Elemental analyses were determined by using a Perkin-Elmer 240c elemental analysis instrument. X-Ray crystallographic analysis was performed with a Siemens SMART CCD and a Siemens P4 diffractometer.

General procedure for the synthesis of compound 4 with microwave irradiation. In a 10 mL Emrys™ reaction vial, an aldehyde **1** (1 mmol), dimedone or 1,3-cyclohexanedione **2** (2 mmol), primary arylamines **3** (1 mmol) and water (1.0 mL)

Table 1
Physical data of compounds **4**.

Entry	Product	Ar	2	Ar'-NH ₂	Time (min)	Mp (°C)	Yield (%)
1	4a	4-Chlorophenyl	2a	<i>p</i> -Toluidine	7	283–285	96
2	4b	4-Bromophenyl	2a	<i>p</i> -Toluidine	8	277–278	93
3	4c	4-Nitrophenyl	2a	<i>p</i> -Toluidine	8	>300	87
4	4d	4-Methoxyphenyl	2a	<i>p</i> -Toluidine	10	241–243	90
5	4e	Benzo[d][1,3]dioxol-5-yl	2a	<i>p</i> -Toluidine	10	263–264	92
6	4f	4-Chlorophenyl	2a	4-Aminophenol	8	>300	90
7	4g	4-Bromophenyl	2a	4-Aminophenol	8	>300	88
8	4h	4-Chlorophenyl	2a	4-Chlorobenzeneamine	8	>300	89
9	4i	4-Bromophenyl	2a	4-Chlorobenzeneamine	8	>300	87
10	4j	4-Nitrophenyl	2a	4-Chlorobenzeneamine	8	>300	85
11	4k	Benzo[d][1,3]dioxol-5-yl	2a	4-Chlorobenzeneamine	10	287–288	88
12	4l	4-Methoxyphenyl	2a	4-Chlorobenzeneamine	8	269–270	87
13	4m	4-Bromophenyl	2a	Aniline	8	245–247	92
14	4n	4-Fluorophenyl	2b	<i>p</i> -Toluidine	8	267–269	95
15	4o	Benzo[d][1,3]dioxol-5-yl	2b	<i>p</i> -Toluidine	8	236–238	89
16	4p	4-Fluorophenyl	2b	<i>p</i> -Toluidine	8	263–264	93
17	4q	4-Bromophenyl	2b	<i>p</i> -Toluidine	9	>300	89
18	4r	4-Nitrophenyl	2b	<i>p</i> -Toluidine	8	>300	87
19	4s	4-Methoxyphenyl	2b	<i>p</i> -Toluidine	8	261–262	88
20	4t	4-Chlorophenyl	2b	4-Aminophenol	8	>300	92
21	4u	4-Methoxyphenyl	2b	4-Chlorobenzeneamine	8	270–272	86
22	4v	4-Fluorophenyl	2b	4-Chlorobenzeneamine	8	298–300	91
23	4w	4-Chlorophenyl	2b	4-Chlorobenzeneamine	8	255–257	89
24	4x	4-Chlorophenyl	2b	Aniline	8	288–290	92
25	4y	4-Methoxyphenyl	2b	Aniline	8	290–291	87

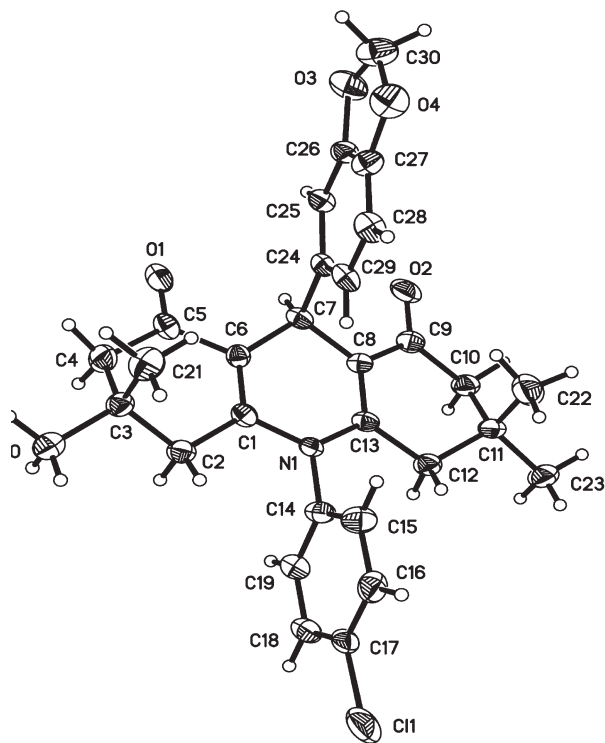


Figure 1. Molecular structure of **4k**.

was mixed and then capped. The mixture was irradiated for a given time at power of 200 W at 140°C. Upon completion, monitored by TLC, the reaction mixture was cooled to room temperature. The solid product was filtered to give the crude product, which was further purified by recrystallization from EtOH (95%).

9-(4-Chlorophenyl)-10-(p-tolyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4a). mp 283–285°C. (lit. mp: 265–267°C) [19]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3058, 2958, 2888, 1639, 1575, 1511, 1486, 1450, 1361, 1278, 1221, 1144, 1089, 841. ¹H NMR: 7.41 (d, 2H, ArH, *J* = 8.4 Hz), 7.29–7.31 (m, 6H, ArH), 5.02 (s, 1H, CH), 2.50 (s, 3H, CH₃), 2.17–2.22 (m, 4H, 2CH₂), 2.00 (d, 2H, CH₂, *J* = 16.0 Hz), 1.77 (d, 2H, CH₂, *J* = 17.2 Hz), 0.88 (s, 6H, CH₃), 0.71 (s, 6H, CH₃). Anal. calcd. for C₃₀H₃₂ClNO₂: C, 76.01; H, 6.80; N, 2.95. Found: C, 76.25; H, 6.71; N, 3.02.

9-(4-Bromophenyl)-10-(p-tolyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4b). mp 277–278°C. (lit. mp: 265–267°C) [20]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3035, 2957, 2870, 1640, 1576, 1511, 1471, 1362, 1278, 1222, 1145, 1069, 842. ¹H NMR: 7.40–7.45 (m, 4H, ArH), 7.25–7.32 (m, 4H, ArH), 5.00 (s, 1H, CH), 2.42 (s, 3H, CH₃), 2.17–2.22 (m, 4H, 2CH₂), 1.99 (d, 2H, CH₂, *J* = 16.0 Hz), 1.77 (d, 2H, CH₂, *J* = 16.0 Hz), 0.88 (s, 6H, CH₃), 0.71 (s, 6H, CH₃). Anal. calcd. for C₃₀H₃₂BrNO₂: C, 69.50; H, 6.22; N, 2.70. Found: C, 69.39; H, 6.13; N, 2.82.

9-(4-Nitrophenyl)-10-(4-tolyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4c). mp >300°C. This

compound was obtained according to earlier general procedure. IR (potassium bromide): 2959, 2930, 1639, 1573, 1512, 1343, 1222, 1146, 834. ¹H NMR: 8.14–8.16 (m, 2H, ArH), 7.57–7.59 (m, 2H, ArH), 7.42 (d, 2H, ArH, *J* = 6.8), 7.35 (d, 2H, ArH, *J* = 7.6), 5.14 (s, 1H, CH), 2.43 (s, 3H, CH₃), 2.19–2.24 (m, 4H, 2CH₂), 2.00 (d, 2H, CH₂, *J* = 16.0 Hz), 1.80 (d, 2H, CH₂, *J* = 17.2 Hz), 0.88 (s, 6H, CH₃), 0.70 (s, 6H, CH₃). Anal. calcd. for C₃₀H₃₂N₂O₄: C, 74.36; H, 6.66; N, 5.78. Found: C, 74.16; H, 6.79; N, 5.73.

9-(4-Methoxyphenyl)-10-(4-tolyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4d). mp: 281–283°C. (lit. mp: 285–287°C) [20]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3039, 2947, 2865, 1572, 1484, 1359, 1175, 1010, 837. ¹H NMR: 7.41 (d, 3H, ArH, *J* = 8.0 Hz), 7.20 (d, 3H, ArH, *J* = 8.8 Hz), 7.80 (d, 2H, ArH, *J* = 8.4 Hz), 4.98 (s, 1H, CH), 3.69 (s, 3H, OCH₃), 2.43 (s, 3H, CH₃), 2.16–2.22 (m, 4H, 2CH₂), 1.99 (d, 2H, CH₂, *J* = 16.0 Hz), 1.76 (d, 2H, CH₂, *J* = 17.2 Hz), 0.88 (s, 6H, CH₃), 0.72 (s, 6H, CH₃). Anal. calcd. for C₃₁H₃₅NO₃: C, 79.28; H, 7.51; N, 2.98. Found: C, 79.47; H, 7.62; N, 2.79.

9-(Benzo[d][1,3]dioxo-5-yl)-10-(p-tolyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4e). mp 263–264°C. (lit. mp: 272–274°C) [20]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 2954, 2871, 1640, 1576, 1486, 1420, 1361, 1278, 1254, 1142, 1042, 813. ¹H NMR: 7.16–7.21 (m, 4H, ArH), 6.72–6.83 (m, 3H, ArH), 5.94 (s, 2H, CH₂), 4.96 (s, 1H, CH), 2.50 (s, 3H, CH₃), 2.16–2.20 (m, 4H, 2CH₂), 2.02 (d, 2H, CH₂, *J* = 16.0 Hz), 1.78 (d, 2H, CH₂, *J* = 17.2 Hz), 0.88 (s, 6H, CH₃), 0.74 (s, 6H, CH₃). Anal. calcd. for C₃₁H₃₃NO₄: C, 76.99; H, 6.88; N, 2.90. Found: C, 76.78; H, 6.98; N, 2.69.

9-(4-Chlorophenyl)-10-(4-hydroxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4f). mp >300°C. (lit. mp: >300°C) [17]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3262, 2960, 2875, 1642, 1640, 1566, 1515, 1451, 1364, 1316, 1264, 1225, 1177, 1145, 1090, 1013, 886, 852, 782. ¹H NMR: 10.0 (s, 1H, OH), 6.92–7.30 (m, 8H, ArH), 5.01 (s, 1H, CH), 2.17–2.22 (m, 4H, 2CH₂), 1.83–2.00 (m, 4H, 2CH₂), 0.90 (s, 6H, CH₃), 0.72 (s, 6H, CH₃). Anal. calcd. for C₂₉H₃₀ClNO₃: C, 73.17; H, 6.35; N, 2.94. Found: C, 73.32; H, 6.21; N, 3.02.

9-(4-Bromophenyl)-10-(4-hydroxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4g). mp >300°C. (lit. mp: >300°C) [17]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3162, 2958, 2875, 1638, 1564, 1513, 1463, 1365, 1316, 1264, 1178, 1146, 1070, 1009, 887, 850, 734, 779. ¹H NMR: 10.0 (s, 1H, OH), 7.42 (d, 2H, ArH, *J* = 8.0 Hz), 7.24 (d, 2H, ArH, *J* = 8.0 Hz), 6.90–7.22 (m, 4H, ArH), 5.1 (s, 1H, CH), 2.16–2.23 (m, 4H, 2CH₂), 1.83–2.00 (m, 4H, 2CH₂), 0.90 (s, 6H, CH₃), 0.72 (s, 6H, CH₃). Anal. calcd. for C₂₉H₃₀BrNO₃: C, 66.92; H, 5.81; N, 2.69. Found: C, 67.08; H, 5.65; N, 2.54.

9-(4-Chlorophenyl)-10-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4h). mp >300°C. (lit. mp: 284–286°C) [20]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 2958, 2870, 1639, 1578, 1490, 1361, 1361, 1263, 1221, 1145, 1090, 1014, 840, 739. ¹H NMR: 7.68 (d, 2H, ArH, *J* = 12.0 Hz), 7.47–7.49 (m, 2H, ArH), 7.28–7.33 (m, 4H, ArH), 5.01 (s, 1H, CH), 2.17–2.22 (m, 4H, 2CH₂), 2.01

(d, 2H, CH₂, *J* = 16.0 Hz), 1.77 (d, 2H, CH₂, *J* = 16.0 Hz), 0.89 (s, 6H, CH₃), 0.72 (s, 6H, CH₃). Anal calcd. for C₂₉H₂₉Cl₂NO₂: C, 70.44; H, 5.91; N, 2.83. Found: C, 70.21; H, 6.02; N, 2.71.

9-(4-Bromophenyl)-10-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4i). mp: 254–256°C. (lit. mp: 249–251°C) [20]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 2958, 2869, 1639, 1578, 1577, 1491, 1361, 1221, 1145, 1089, 1009, 839. ¹H NMR: 7.68 (d, 2H, ArH, *J* = 8.0 Hz), 7.42–7.48 (m, 4H, ArH), 7.26 (d, 2H, ArH, *J* = 8.0 Hz), 4.99 (s, 1H, CH), 2.17–2.23 (m, 4H, 2CH₂), 2.01 (d, 2H, CH₂, *J* = 16.0 Hz), 1.76 (d, 2H, CH₂, *J* = 16.0 Hz), 0.89 (s, 6H, CH₃), 0.72 (s, 6H, CH₃). Anal calcd. for C₂₉H₂₉BrClNO₂: C, 64.63; H, 5.42; N, 2.60. Found: C, 64.81; H, 5.28; N, 2.68.

9-(4-Nitrophenyl)-10-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4j). mp >300°C. This compound was obtained according to earlier general procedure. IR (potassium bromide): 2959, 2870, 1638, 1577, 1491, 1361, 1342, 1222, 1145, 1089, 1009, 831. ¹H NMR: 7.67–7.70 (m, 2H, ArH), 7.48–7.50 (m, 2H, ArH), 7.28–7.33 (m, 4H, ArH), 5.01 (s, 1H, CH), 2.17–2.22 (m, 4H, 2CH₂), 2.02 (d, 2H, CH₂, *J* = 16.0 Hz), 1.77 (d, 2H, CH₂, *J* = 16.0 Hz), 0.89 (s, 6H, CH₃), 0.72 (s, 6H, CH₃). Anal calcd. for C₂₉H₂₉ClN₂O₄: C, 68.97; H, 5.79; N, 5.55. Found: C, 69.14; H, 5.61; N, 5.42.

9-(Benzo[d][1,3]dioxo-5-yl)-10-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4k). mp 287–288°C. (lit. mp: 287–288°C) [24]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 2955, 2871, 1641, 1578, 1490, 1360, 1254, 1221, 1141, 1042, 921, 813. ¹H NMR: 7.68 (d, 2H, ArH, *J* = 8.8 Hz), 7.42–7.45 (m, 2H, ArH), 6.77–6.79 (m, 3H, ArH), 5.94 (s, 2H, CH₂), 4.96 (s, 1H, CH), 2.16–2.20 (m, 4H, 2CH₂), 2.03 (d, 2H, CH₂, *J* = 17.6 Hz), 1.78 (d, 2H, CH₂, *J* = 17.2 Hz), 0.89 (s, 6H, CH₃), 0.75 (s, 6H, CH₃). Anal calcd. for C₃₀H₃₀ClNO₄: C, 71.49; H, 6.00; N, 2.78. Found: C, 71.23; H, 6.18; N, 2.69.

9-(4-Methoxyphenyl)-10-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4l). mp 269–270°C. (lit. mp: 269–271°C) [27]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 2952, 1641, 1578, 1491, 1361, 1259, 1173, 1140, 998, 834, 740. ¹H NMR: 7.68 (d, 2H, ArH, *J* = 8.0), 7.44–7.47 (m, 2H, ArH), 7.21 (d, 2H, ArH, *J* = 8.0 Hz), 6.80 (d, 2H, ArH, *J* = 8.0 Hz), 4.97 (s, 1H, CH), 3.69 (s, 3H, OCH₃), 2.16–2.22 (m, 4H, 2CH₂), 2.00 (d, 2H, CH₂, *J* = 16.0 Hz), 1.77 (d, 2H, CH₂, *J* = 17.6 Hz), 0.89 (s, 6H, CH₃), 0.73 (s, 6H, CH₃). Anal calcd. for C₃₀H₃₂ClNO₃: C, 73.53; H, 6.58; N, 2.86. Found: C, 73.73; H, 6.62; N, 2.71.

9-(4-Bromophenyl)-10-phenyl-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4m). mp 285–287°C. (lit. mp: > 300°C) [20]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3060, 2956, 2869, 1639, 1577, 1491, 1452, 1361, 1261, 1176, 1008, 838. ¹H NMR: 7.55–7.64 (m, 3H, ArH), 7.45 (d, 4H, ArH, *J* = 8.0 Hz), 7.27 (d, 2H, ArH, *J* = 8.0 Hz), 5.01 (s, 1H, CH), 2.18–2.23 (m, 4H, 2CH₂), 2.01 (d, 2H, CH₂, *J* = 16.0 Hz), 1.75 (d, 2H, CH₂, *J* = 16.0 Hz), 0.87 (s, 6H, CH₃), 0.71 (s, 6H, CH₃). Anal calcd. for C₂₉H₃₀BrNO₂: C, 69.05; H, 5.99; N, 2.78. Found: 69.18; H, 5.76; N, 2.54.

9-(4-Fluorophenyl)-10-(4-tolyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4n). mp 267–269°C. (lit. mp: 262–294°C) [17]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3063, 2931, 2870, 1651, 1575, 1508, 1451, 1142, 1000, 843. ¹H NMR: 7.41 (d, 2H, ArH, *J* = 7.6 Hz), 7.30–7.34 (m, 4H, ArH), 7.04–7.08 (m, 2H, ArH), 5.03 (s, 1H, CH), 2.42 (s, 3H, CH₃), 2.20–2.22 (m, 4H, 2CH₂), 2.00 (d, 2H, CH₂, *J* = 16.0 Hz), 1.77 (d, 2H, CH₂, *J* = 17.6 Hz), 0.88 (s, 6H, CH₃), 0.71 (s, 6H, CH₃). Anal calcd. for C₃₀H₃₂FNO₂: C, 78.75; H, 7.05; N, 3.06. Found: C, 78.94; H, 7.18; N, 3.02.

9-(Benzo[d][1,3]dioxo-5-yl)-10-(4-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4o). mp 236–238°C. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3031, 2945, 2888, 1642, 1570, 1486, 1362, 1285, 1231, 1135, 1040, 857, 799. ¹H NMR: 7.37 (d, 3H, ArH, *J* = 8.0 Hz), 7.16–7.18 (m, 1H, ArH), 6.72–6.79 (m, 3H, ArH), 5.94 (s, 2H, CH₂), 5.06 (s, 1H, CH), 2.97 (s, 3H, CH₃), 2.18–2.24 (m, 6H, 3CH₂), 1.91–1.96 (m, 2H, CH₂), 1.76–1.84 (m, 2H, CH₂), 1.59–1.65 (m, 2H, CH₂). Anal calcd. for C₂₇H₂₅NO₄: C, 75.86; H, 5.89; N, 3.28. Found: 75.81; H, 6.01; N, 3.41.

9-(4-Fluorophenyl)-10-(p-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4p). mp 263–264°C. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3059, 2929, 2870, 1633, 1571, 1507, 1360, 1284, 1231, 1134, 839. ¹H NMR: 7.37–7.39 (m, 3H, ArH), 7.19–7.31 (m, 3H, ArH), 7.02–7.07 (m, 2H, ArH), 5.13 (s, 1H, CH), 2.40 (s, 3H, CH₃), 2.18–2.22 (m, 6H, 3CH₂), 1.92–1.97 (m, 2H, CH₂), 1.79–1.84 (m, 2H, CH₂), 1.63–1.65 (m, 2H, CH₂). Anal calcd. for C₂₆H₂₄FNO₂: C, 77.78; H, 6.03; N, 3.49. Found: C, 77.94; H, 5.85; N, 3.31.

9-(4-Bromophenyl)-10-(p-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4q). mp >300°C. (lit. mp: > 300°C) [17]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3040, 2923, 2867, 1644, 1572, 1510, 1485, 1361, 1283, 1230, 1134, 1069, 831. ¹H NMR: 7.37–7.43 (m, 5H, ArH), 7.24 (d, 2H, ArH, *J* = 8.4 Hz), 7.14 (d, 1H, ArH, *J* = 8.4 Hz), 5.10 (s, 1H, CH), 2.40 (s, 3H, CH₃), 2.18–2.21 (m, 8H, 4CH₂), 1.92–1.97 (m, 2H, CH₂), 1.77–1.85 (m, 2H, CH₂). Anal calcd. for C₂₆H₂₄BrNO₂: C, 67.54; H, 5.23; N, 3.03. Found: C, 67.72; H, 5.06; N, 3.19.

9-(4-Nitrophenyl)-10-(p-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4r). mp >300°C. (lit. mp: > 300°C) [17]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 2953, 2915, 1631, 1600, 1574, 1511, 1342, 1284, 1230, 1179, 1132. ¹H NMR: 7.31–8.13 (m, 8H, ArH), 5.23 (s, 1H, CH), 2.40 (s, 3H, CH₃), 1.94–2.89 (m, 8H, 4CH₂), 1.79–1.85 (m, 2H, CH₂), 1.62–1.65 (m, 2H, CH₂). Anal calcd. for C₂₆H₂₄N₂O₄: C, 72.88; H, 5.65; N, 6.54. Found: C, 72.72; H, 5.84; N, 6.70.

9-(4-Methoxyphenyl)-10-(p-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4s). mp 241.2–243.0°C. (lit. mp: 256–257°C) [17]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 2938, 1637, 1569, 1509, 1360, 1287, 1232, 1181, 1130, 954, 913, 825, 758. ¹H NMR: 7.37–7.86 (m, 4H, ArH), 7.18 (d, 2H, ArH, *J* = 8.8 Hz), 6.80 (d, 2H, ArH, *J* = 8.4 Hz), 5.07 (s, 1H, CH), 3.70 (s, 3H, OCH₃), 2.40 (s, 3H, CH₃), 2.18–2.25 (m, 6H, 3CH₂), 1.80–1.83 (m, 4H, 2CH₂), 1.59–1.61 (m, 2H, CH₂). Anal calcd. for C₂₇H₂₇NO₃: C, 78.42; H, 6.58; N, 3.39. Found: C, 78.69; H, 6.72; N, 3.12.

9-(4-Chlorophenyl)-10-(4-hydroxyphenyl)-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4t). mp >300°C. (lit. mp: >300°C) [17]. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3161, 1636, 1561, 1489, 1453, 1385, 1362, 1269, 1234, 1140, 1088, 959. ¹H NMR: 9.94 (s, 1H, OH), 6.89–7.28 (m, 8H, ArH), 5.11 (s, 1H, CH), 2.17–2.25 (m, 6H, 3CH₂), 1.96–2.04 (m, 2H, CH₂), 1.80–1.86 (m, 2H, CH₂), 1.59–1.67 (m, 2H, CH₂). Anal calcd. for C₂₅H₂₂ClNO₃: C, 71.51; H, 5.28; N, 3.34. Found: C, 71.38; H, 5.32; N, 3.40.

9-(4-Methoxyphenyl)-10-(4-chlorophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4u). mp 270–272°C. This compound was obtained according to earlier general procedure. IR (potassium bromide): 2938, 1634, 1569, 1509, 1363, 1284, 1230, 1181, 1132, 955, 846, 756. ¹H NMR: 7.68 (d, 2H, ArH, *J* = 12.0), 7.45–7.47 (m, 2H, ArH), 7.21 (d, 2H, ArH, *J* = 8.0 Hz), 6.80 (d, 2H, ArH, *J* = 8.0 Hz), 4.97 (s, 1H, CH), 3.69 (s, 3H, OCH₃), 2.19–2.23 (m, 6H, 3CH₂), 1.82–1.84 (m, 4H, 2CH₂), 1.59–1.62 (m, 2H, CH₂). Anal calcd. for C₂₆H₂₄ClNO₃: C, 71.97; H, 5.57; N, 3.23. Found: C, 72.12; H, 5.38; N, 3.32.

9-(4-fluorophenyl)-10-(4-chlorophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4v). mp 298–299°C. This compound was obtained according to earlier general procedure. IR (potassium bromide): 2962, 1634, 1572, 1505, 1360, 1283, 1232, 1182, 1132, 956, 839, 759. ¹H NMR: 7.65 (d, 2H, ArH, *J* = 8.0), 7.29–7.33 (m, 3H, ArH), 7.02–7.06 (m, 3H, ArH), 5.12 (s, 1H, CH), 2.19–2.24 (m, 6H, 3CH₂), 1.92–1.97 (m, 2H, CH₂), 1.80–1.84 (m, 2H, CH₂), 1.62–1.65 (m, 2H, CH₂). Anal calcd. for C₂₅H₂₁ClFNO₂: C, 71.17; H, 5.02; N, 3.32. Found: C, 71.02; H, 5.18; N, 3.21.

9-(4-Chlorophenyl)-10-(4-chlorophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4w). mp 255–257°C. This compound was obtained according to earlier general procedure. IR (potassium bromide): 2946, 1633, 1571, 1489, 1362, 1284, 1183, 1134, 1089, 858, 821, 757. ¹H NMR: 7.63–7.66 (m, 2H, ArH), 7.53–7.57 (m, 2H, ArH), 7.17–7.20 (m, 2H, ArH), 6.79 (d, 2H, ArH, *J* = 8.0), 5.06 (s, 1H, CH), 2.18–2.26 (m, 6H, 3CH₂), 1.91–1.97 (m, 2H, CH₂), 1.80–1.85 (m, 2H, CH₂), 1.59–1.65 (m, 2H, CH₂). Anal calcd. for C₂₅H₂₁Cl₂NO₂: C, 68.50; H, 4.83; N, 3.20. Found: C, 68.38; H, 4.99; N, 3.07.

9-(4-Chlorophenyl)-10-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4x). mp 288–290°C. This compound was obtained according to earlier general procedure. IR (potassium bromide): 3044, 2953, 1637, 1570, 1489, 1425, 1360, 1269, 1281, 1137, 1088, 956, 835. ¹H NMR: 8.753–7.60 (m, 3H, ArH), 7.19–7.33 (m, 6H, ArH), 5.13 (s, 1H, CH), 2.19–2.25 (m, 6H, 3CH₂), 1.90–1.95 (m, 2H, CH₂), 1.79–1.81 (m, 2H, CH₂), 1.60–1.63 (m, 2H, CH₂). Anal calcd. for C₂₅H₂₂ClNO₂: C, 74.34; H, 5.49; N, 3.47. Found: C, 74.18; H, 5.62; N, 3.31.

9-(4-Methoxyphenyl)-10-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8-[2H,5H]-dione (4y). mp 290–291°C. (lit. mp: 270–272°C) [28]. IR (potassium bromide): 2943, 2887, 1635, 1569, 1509, 1359, 1284, 1229, 1181, 1132, 953, 857. ¹H NMR: 7.55–7.57 (m, 4H, ArH), 7.30–7.35 (m, 1H, ArH), 7.18–7.21 (m, 2H, ArH), 6.79–7.82 (m, 2H, ArH), 5.09 (s, 1H, CH), 3.70 (s, 3H, OCH₃), 2.19–2.24 (m, 6H, 3CH₂), 1.84–1.90 (m, 2H, CH₂), 1.79–1.81 (m, 2H, CH₂), 1.60–1.64 (m, 2H, CH₂). Anal calcd. for C₂₆H₂₅NO₃: C, 78.17; H, 6.31; N, 3.51. Found: C, 78.01; H, 6.52; N, 3.58.

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