Microwave-Assisted Multicomponent Reaction for the Synthesis of New and Significative Bisfunctional Compounds Containing Two Furo[3,4-b]quinoline and Acridinedione Skeletons

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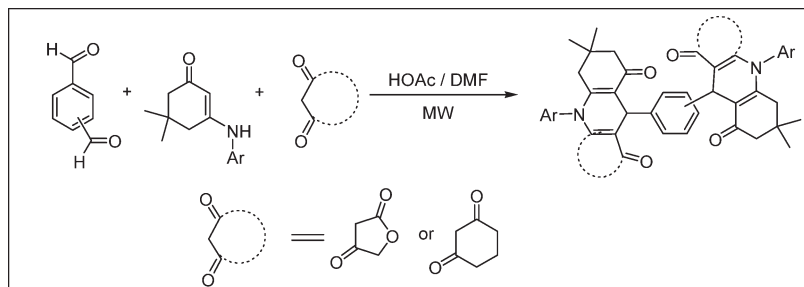
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In this study, a series of new and significative bisfunctional compounds containing two furo[3,4-b]quinoline and acridinedione skeletons has been synthesized through a rapid one-pot three-component reaction of dialdehydes with N-aryl enaminones and cyclic-1,3-dicarbonyl compounds (tetronic acid and cyclohexane-1,3-dione) in mixed solvent of glacial acetic acid and N,N-dimethylformamide under microwave irradiation without catalyst. This method has the advantages of good yield and simple workup procedure.

INTRODUCTION

Podophyllotoxin **1** (Figure 1) is an important lignan that inhibits microtubule assembly [1]. However, because attempts to use it for the treatment of human neoplasia were mostly unsuccessful and complicated by side effects, extensive structural modifications have been performed to obtain more potent and less toxic anti-cancer agents [2]. Among them, 4-aza podophyllotoxin derivative **2** (Fig. 1) has been reported to be a powerful DNA topoisomerase inhibitor, working through a mechanism of action entirely different from that of the parent, natural podophyllotoxin [3,4]. This suggests that substitution of the carbon atom at position 4 of podophyllotoxin by a nitrogen atom would bring about great changes in the biological profile. In view of the important biological properties of the azapodophyllotoxin, the modifications on the scaffold of aza-analogs may also bring significant changes in pharmacological activities.

The furoquinolines **A** as a kind of azapodophyllotoxin derivative have been synthesized by the reactions of single aldehydes with equivalent of appropriate cyclic-1,3-dicarbonyl compounds and N-aryl enaminones in our previous communications [5]. In addition, with a 1,4-dihydropyridine (1,4-DHP) parent nucleus, acridine-1,8-diones **B**, which showed interesting physical properties

such as photoinitiators [6] have been prepared [7]. However, all these compounds contain only single furoquinoline skeleton or acridinedione unit. To the best of our knowledge, the compounds of type **C** and **D** including two furoquinoline skeletons or acridinedione units have been seldom reported (Fig. 1). With the aim to broaden the diversity of heterocyclic compound library and in continuation of our recent interest in the construction of heterocyclic scaffolds [8], we developed a facile, three-component reaction between dialdehydes, N-aryl enaminones, and cyclic-1,3-dicarbonyl compounds (tetronic acid and 1,3-cyclohexanedione) under microwave (MW) heating to afford a series of new polycyclic fused compounds **C** and **D**, including two furoquinoline skeletons or acridinedione units, respectively (Scheme 1).

RESULTS AND DISCUSSION

Enaminones and related compounds possessing the structural unit (N—C=C—Z, Z=COR, CO2R, *etc.*) are versatile synthetic intermediates in organic chemistry that combine the ambient nucleophilicity of enamine and the electrophilicity of enones [9]. They are frequently applied in the preparation of heterocycles [10]. Our strategy of synthesizing the bisfunctional
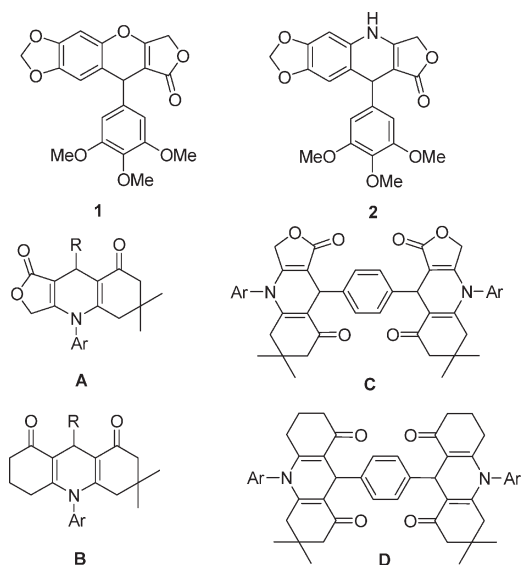
Figure 1. The podophyllotoxin derivatives and the unsymmetrical acridinediones.

Scheme 2

2a: Ar = 4-ClC$_6$H$_4$ **2b**: Ar = C$_6$H$_5$
2c: Ar = 4-CH$_3$C$_6$H$_4$ **2d**: Ar = 4-OHC$_6$H$_4$

compounds, furoquinolines of type **C** and acridinediones of type **D**, was through the reaction of a dialdehyde with a appropriate cyclic-1,3-dicarbonyl derivative and a preformed *N*-aryl enaminones. Representing aromatic amines with electron-rich group, 4-methylphenylamine electron-withdrawing group, 4-chlorobenzenamine, and 4-aminophenol, and phenylamine were selected for our study. The preparation of enaminones **2a–d** was commonly achieved by refluxing the reaction mixture in an aromatic solvent, with the removal of the produced water by azeotropic distillation [11]. We found that enaminones (**2a–d**) could be obtained in good to excellent yields by MW heating the mixture of the corresponding amine and 5,5-dimethyl-1,3-cyclohexanedione in EtOH (95%) at 100°C for 4–6 min (Scheme 2, Table 1) [5].

Choosing an appropriate solvent is of crucial importance for the successful microwave-assisted synthesis. To search for the optimal solvent, the microwave-assisted reaction of equimolar amount of terephthalaldehyde (**1a**) with 3-(*p*-tolylamino)-5,5-dimethylcyclohex-2-enone (**2c**) and tetronic acid (**3a**) was examined using
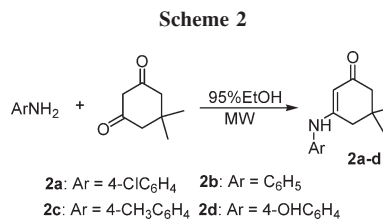
ethylene glycol, glacial acetic acid (HOAc), *N,N*-dimethylformamide (DMF), and mixed solvent of glacial acetic acid and DMF as solvent at 100°C, respectively. All the reactions were carried out under microwave irradiation (initial power 100 W and maximum power 250 W; Table 2).

As shown in Table 2, the reactions using the mixed solvent (V_{HOAc}/V_{DMF}: 2:1) as the solvent resulted in higher yields and shorter reaction time than those using ethylene glycol, HOAc, DMF, and other mixed solvent as solvents. Thus, the mixed solvent (V_{HOAc}/V_{DMF}: 2:1) was used as the solvent for further optimization of reaction conditions, the same reaction was carried out at temperatures ranging from 80 to 140°C, with an increment of 10°C each time. The yield of product **4c** was increased, and the reaction time was shortened when the temperature was increased from 80 to 120°C. The yield leveled off when the temperature was further increased to 130 and 140°C. Therefore, the temperature of 120°C was chosen for all further microwave-assisted reactions (Table 3).

Under these optimal conditions [the mixed solvent (V_{HOAc}/V_{DMF}: 2:1, 2.0 mL), 120°C], the reactions of different dialdehydes, various *N*-aryl enaminones, and tetronic acid were performed. Initially, to test the scope of *N*-aryl enaminones substrates, dialdehydes and tetronic acid were used as model substrates (Table 4, entries 1–6), and the results indicated that *N*-aryl enaminones bearing functional groups such as chloro or methyl are suitable for the reaction. At the same time, we have also observed delicate electronic effects, that is, *N*-aryl enaminones with electron-rich groups (Table 4, entries 3 and 6) reacted rapidly, while electron-withdrawing groups on the benzene ring (Table 4, entries 1 and 4) decreased the reactivity, requiring longer reaction times.

Scheme 1
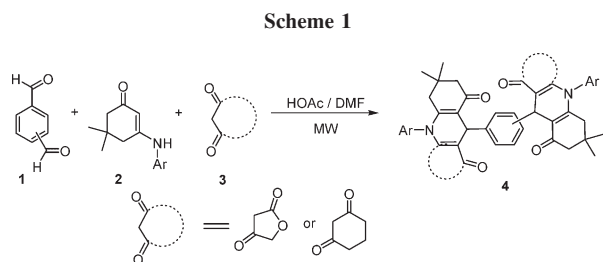
Table 1

Reaction times and yields for enaminones **2a–2d**.

Entry	Product	Time (min)	Yield (%)	m.p. (°C)
1	**2a**	4	89	207–209
2	**2b**	5	91	189–190
3	**2c**	5	93	206–208
4	**2d**	6	94	249–251

Table 2

Solvent optimization for the synthesis of **4c** under MW.

Entry	Solvent	Time (min)	Yield (%)
1	Ethylene glycol	14	80
2	HOAc	14	81
3	DMF	15	78
4	HOAc/DMF (3:1)	12	83
5	HOAc/DMF (2:1)	12	85
6	HOAc/DMF (1:1)	12	82

Table 3

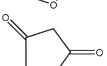Temperature optimization for the synthesis of **4c** under MW.
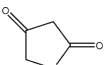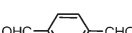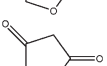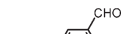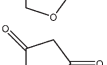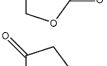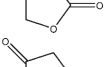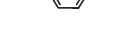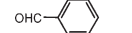
Entry	Temp (°C)	Time (min)	Yield (%)
1	80	18	70
2	90	16	81
3	100	12	85
4	110	10	89
5	120	8	92
6	130	8	90
7	140	6	86

To further expand the scope of this method, the replacement of tetronic acid (**3a**) with 1,3-cyclohexane-dione (**3b**) was examined. To our delight, under the optimized conditions described earlier, the reactions proceeded smoothly as well (Table 4, entries 7–11). The new polycyclic-fused compounds including two acridinedione units were obtained without byproduct.

Additionally, to compare with microwave-assisted reactions, the same temperature and time were applied

to synthesize the same product under classical heating (CH) conditions. The results listed in Table 4 show the specific activation of this reaction by MW heating. Simultaneously, the reaction yields were obviously increased. The difference in yields (MW > CH) may be a consequence of both thermal effects and specific effects induced by the microwave field [12,13]. The reactants in these multicomponent reactions (MCRs)

Table 4

The synthesis of compounds **4** at 120°C.

Entry	4	1	2	3	Time (min)	Yield[a] (%)	m.p. (°C)
1	**4a**	OHC—⬡—CHO	**2a**		14	85 (31)	>300
2	**4b**	OHC—⬡—CHO	**2b**		10	89 (44)	>300
3	**4c**	OHC—⬡—CHO	**2c**		8	92 (40)	>300
4	**4d**	OHC—⬡—CHO	**2a**		16	82 (32)	>300
5	**4e**	OHC—⬡—CHO	**2b**		10	86 (29)	>300
6	**4f**	OHC—⬡—CHO	**2c**		10	88 (35)	>300
7	**4g**	OHC—⬡—CHO	**2a**		12	84 (51)	>300
8	**4h**	OHC—⬡—CHO	**2c**		8	92 (46)	>300
9	**4i**	OHC—⬡—CHO	**2d**		7	87 (41)	>300
10	**4j**	OHC—⬡—CHO	**2c**		13	85 (38)	>300
11	**4k**	OHC—⬡—CHO	**2d**		10	90 (45)	>300

[a] Isolated yields under classical heating (CH) conditions.
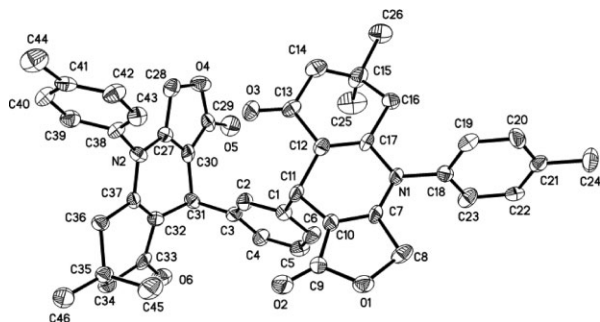
Figure 2. ORTEP diagram of **4f**.

contain dipoles and proceed via relatively polar intermediates, which enhance their interactions with MW and consequently benefit significantly from MW irradiation with respect to more efficient yield and product purity.

The mechanism of these reactions is similar to those we have previously reported [5,7], which includes sequential condensation, addition, cyclization, and elimination.

In this study, all the products were characterized by melting point, IR, ^{1}H NMR spectral data, and elemental analysis. Furthermore, the structure of **4f** was established by X-ray crystallographic analysis (Fig. 2) [14].

In summary, we have successfully combined the advantages of microwave technology with multicomponent reactions to facilitate the rapid construction of bisfuro[3,4-*b*]quinoline and bisacridinedione skeletons from readily obtainable and inexpensive materials. Particularly, valuable features of this method included the good to excellent yields and operational simplicity as well as increased safety for small-scale high-speed synthesis. In addition, this series of bisfunctional compounds containing two furo[3,4-*b*]quinoline and acridinedione skeletons may prove new classes of biologically active compounds for biomedical screening, which is in progress in our laboratory.

EXPERIMENTAL

Microwave irradiation was carried out with a microwave oven Emrys^{TM} Creator from Personal Chemistry, Uppsala, Sweden. Melting points were determined in the open capillaries and were uncorrected. IR spectra were taken on a FTIR-Tensor 27 spectrometer in KBr pellets and reported in cm^{-1}. ^{1}H NMR spectra were measured on a Bruker DPX 400 MHz spectrometer using TMS as an internal standard and DMSO-d_6 as solvent. Elemental analysis was determined by using a Perkin-Elmer 240c elemental analysis instrument. HRMS (ESI) was determined by using micrOTOF-QII HRMS instrument (BRUKER). X-ray crystallographic analysis was performed with a Siemens SMART CCD and a Siemens P4 diffractometer.

General procedure for the one-pot synthesis of furo[3,4-*b*]quinoline and acridinedione derivative 4 under microwave irradiation conditions. Typically, in a 10-mL Emrys^{TM} reaction vial, dialdehyde 1 (1 mmol), *N*-aryl enaminones 2 (1 mmol), tetronic acid or 1,3-cyclohexanedione 3 (1 mmol), and HOAc/DMF (2:1, 2 mL) were mixed and then capped. The mixture was irradiated for a given time at 120°C under microwave irradiation (initial power 100 W and maximum power 250 W). Upon completion, monitored by TLC, the reaction mixture was filtered to give the crude product, which was further purified by recrystallization from EtOH (95%) to give pure furo[3,4-*b*]quinoline and acridinedione derivative **4**.

4-(4-Chlorophenyl)-9-(4-(4-(4-chlorophenyl)-1,3,4,5,6,7,8,9-octahydro-6,6-dimethyl-1,8-dioxofuro[3,4-b]quinolin-9-yl)-phenyl)-6,7-dihydro-6,6-dimethylfuro[3,4-b]quinoline-1,8(3H,4H,5H,9H)-dione (4a). This compound was obtained as pale yellow solid (95% ethanol), m.p. > 300°C; IR (potassium bromide, cm^{-1}): 1755 (C=O), 1682 (C=O); ^{1}H NMR: δ 7.68–7.60 (m, 8H, Ar-H), 7.25 (s, 4H, Ar-H), 4.75 (s, 1H, CH), 4.74 (s, 1H, CH), 4.63–4.59 (m, 2H, CH$_2$), 4.56–4.49 (m, 2H, CH$_2$), 2.23–2.09 (m, 8H, 4CH$_2$), 0.94–0.86 (m, 12H, 4CH$_3$). Anal. Calcd. for C$_{44}$H$_{38}$Cl$_2$N$_2$O$_6$: C, 69.38; H, 5.03; N, 3.68. Found: C, 71.08; H, 5.07; N, 3.66.

4-Phenyl-6,7-dihydro-9-(4-(1,3,4,5,6,7,8,9-octahydro-6,6-dimethyl-1,8-dioxo-furo[3,4-b]quinolin-9-yl)phenyl)-6,6-dimethylfuro[3,4-b]quinoline-1,8(3H,4H,5H,9H)-dione (4b). This compound was obtained as pale yellow solid (95% ethanol), m.p. > 300°C; IR (potassium bromide, cm^{-1}): 1756 (C=O), 1680 (C=O); ^{1}H NMR: δ 7.59–7.54 (m, 10H, Ar-H), 7.26 (s, 4H, Ar-H), 4.77 (s, 1H, CH), 4.75 (s, 1H, CH), 4.62–4.58 (m, 2H, CH$_2$), 4.51–4.45 (m, 2H, CH$_2$), 2.23–2.07 (m, 8H, 4CH$_2$), 0.93–0.85 (m, 12H, 4CH$_3$). Anal. Calcd. for C$_{44}$H$_{40}$N$_2$O$_6$: C, 76.28; H, 5.82; N, 4.04. Found: C, 76.41; H, 5.86; N, 4.15.

4-p-Tolyl-6,7-dihydro-9-(4-(1,3,4,5,6,7,8,9-octahydro-6,6-dimethyl-1,8-dioxo-4-p-tolylfuro[3,4-b]quinolin-9-yl)phenyl)-6,6-dimethyl-furo[3,4-b]quinoline-1,8(3H,4H,5H,9H)-dione (4c). This compound was obtained as pale yellow solid (95% ethanol), m.p. > 300°C; IR (potassium bromide, cm^{-1}): 1755 (C=O), 1681 (C=O); ^{1}H NMR: δ 7.44–7.40 (m, 8H, Ar-H), 7.24 (s, 4H, Ar-H), 4.76 (s, 2H, 2CH), 4.58 (d, 2H, J = 16.0 Hz, CH$_2$), 4.49 (d, 2H, J = 16.0 Hz, CH$_2$), 2.40 (s, 6H, 2CH$_3$), 2.21–2.09 (m, 8H, 4CH$_2$), 0.93 (s, 6H, 2CH$_3$), 0.89 (s, 6H, 2CH$_3$). Anal. Calcd. for C$_{46}$H$_{44}$N$_2$O$_6$: C, 76.64; H, 6.15; N, 3.89. Found: C, 76.69; H, 6.14; N, 3.83. HRMS (ESI): *m/z* calcd for: 743.3092 [M+Na]$^+$, found: 743.3099.

4-(4-Chlorophenyl)-9-(3-(4-(4-chlorophenyl)-1,3,4,5,6,7,8,9-octahydro-6,6-dimethyl-1,8-dioxofuro[3,4-b]quinolin-9-yl)phenyl)-6,7-dihydro-6,6-dimethylfuro[3,4-b]quinoline-1,8(3H,4H,5H,9H)-dione (4d). This compound was obtained as pale yellow solid (95% ethanol), m.p. > 300°C; IR (potassium bromide, cm^{-1}): 1756 (C=O), 1681 (C=O); ^{1}H NMR: δ 7.67 (s, 8H, Ar-H), 7.48 (s, 1H, Ar-H), 7.20–7.17 (m, 1H, Ar-H), 7.07 (d, 2H, J = 7.2 Hz, Ar-H), 4.82 (s, 2H, 2CH), 4.68 (d, 2H, J = 16.0 Hz, CH$_2$), 4.55 (d, 2H, J = 16.0 Hz, CH$_2$), 2.18–2.14 (m, 8H, Ar-H), 0.94 (s, 6H, 2CH$_3$), 0.85 (s, 6H, 2CH$_3$). Anal. Calcd. for C$_{44}$H$_{38}$Cl$_2$N$_2$O$_6$: C, 69.38; H, 5.03; N, 3.68. Found: C, 69.45; H, 5.00; N, 3.72.

4-Phenyl-6,7-dihydro-9-(3-(1,3,4,5,6,7,8,9-octahydro-6,6-dimethyl-1,8-dioxo-4-phenylfuro[3,4-b]quinolin-9-yl)phenyl)-6,6-dimethyl-furo[3,4-b]quinoline-1,8(3H,4H,5H,9H)-dione (4e). This compound was obtained as pale yellow solid (95%

ethanol), m.p. > 300°C; IR (potassium bromide, cm^{-1}): 1756 (C=O), 1680 (C=O); ^{1}H NMR: δ 7.60 (s, 10H, Ar-H), 7.49 (s, 1H, Ar-H), 7.21–7.18 (m, 1H, Ar-H), 7.08 (d, 2H, J = 7.6 Hz, Ar-H), 4.83 (s, 2H, 2CH), 4.67 (d, 2H, J = 16.4 Hz, CH$_2$), 4.49 (d, 2H, J = 16.4 Hz, CH$_2$), 2.23–2.09 (m, 8H, 4CH$_2$), 0.92 (s, 6H, 2CH$_3$), 0.84 (s, 6H, 2CH$_3$). Anal. Calcd. for C$_{44}$H$_{40}$N$_2$O$_6$: C, 76.28; H, 5.82; N, 4.04. Found: C, 76.34; H, 5.80; N, 4.08.

4-p-Tolyl-6,7-dihydro-9-(3-(1,3,4,5,6,7,8,9-octahydro-6,6-dimethyl-1,8-dioxo-4-p-tolylfuro[3,4-b]quinolin-9-yl)phenyl)-6,6-dimethyl-furo[3,4-b]quinoline-1,8(3H,4H,5H,9H)-dione (4f). This compound was obtained as pale yellow solid (95% ethanol), m.p. > 300°C; IR (potassium bromide, cm^{-1}): 1753 (C=O), 1679 (C=O); ^{1}H NMR: δ 7.47–7.39 (m, 9H, Ar-H), 7.21–7.17 (m, 1H, Ar-H), 7.06 (d, 2H, J = 7.2 Hz, Ar-H), 4.81 (s, 2H, 2CH), 4.65 (d, 2H, J = 16.0 Hz, CH$_2$), 4.48 (d, 2H, J = 16.4 Hz, CH$_2$), 2.41 (s, 6H, 2CH$_3$), 2.22–2.08 (m, 8H, 4CH$_2$), 0.92 (s, 6H, 2CH$_3$), 0.83 (s, 6H, 2CH$_3$). Anal. Calcd. for C$_{46}$H$_{44}$N$_2$O$_6$: C, 76.64; H, 6.15; N, 3.89. Found: C, 76.34; H,5.99; N, 4.07. HRMS (ESI): m/z calcd for: 743.3092 [M+Na]$^+$, found: 743.3090.

10-(4-Chlorophenyl)-9-(4-(10-(4-chlorophenyl)-1,2,3,4,5,6,7,8,9,10-decahydro-3,3-dimethyl-1,8-dioxoacridin-9-yl)phenyl)-3,4,6,7-tetrahydro-3,3-dimethylacridine-1,8(2H,5H,9H,10H)-dione (4g). This compound was obtained as pale yellow solid (95% ethanol), m.p. > 300°C; IR (potassium bromide, cm^{-1}): 1639 (C=O); ^{1}H NMR: δ 7.68–7.44 (m, 8H, Ar-H), 7.16 (s, 4H, Ar-H), 5.06–5.01 (m, 2H, 2CH), 2.29–2.09 (m, 10H, 5CH$_2$), 2.04–1.92 (m, 4H, 2CH$_2$), 1.83–1.79 (m, 4H, 2CH$_2$), 1.66–1.61 (m, 2H, CH$_2$), 0.89 (s, 6H, 2CH$_3$), 0.70 (s, 6H, 2CH$_3$). Anal. Calcd. for C$_{48}$H$_{46}$Cl$_2$N$_2$O$_4$: C, 73.37; H, 5.90; N, 3.56. Found: C, 75.35; H, 6.00; N, 3.53.

10-p-Tolyl-3,4,6,7-tetrahydro-9-(4-(1,2,3,4,5,6,7,8,9,10-decahydro-3,3-dimethyl-1,8-dioxo-10-p-tolylacridin-9-yl)phenyl)-3,3-dimethyl-acridine-1,8(2H,5H,9H,10H)-dione (4h). This compound was obtained as pale yellow solid (95% ethanol), m.p. > 300°C; IR (potassium bromide, cm^{-1}): 1634 (C=O); ^{1}H NMR: δ 7.41–7.25 (m, 8H, Ar-H), 7.16 (s, 4H, Ar-H), 5.07–5.04 (m, 2H, 2CH), 2.42 (s, 6H, 2CH$_3$), 2.29–2.13 (m, 10H, 5CH$_2$), 2.05–1.92 (m, 4H, 2CH$_2$), 1.83–1.78 (m, 4H, 2CH$_2$), 1.64–1.60 (m, 2H, CH$_2$), 0.87 (s, 6H, 2CH$_3$), 0.69 (s, 6H, 2CH$_3$). Anal. Calcd. for C$_{50}$H$_{52}$N$_2$O$_4$: C, 80.61; H, 7.04; N, 3.76. Found: C, 80.69; H, 7.06; N, 3.73.

10-(4-Hydroxyphenyl)-3,4,6,7-tetrahydro-9-(4-(1,2,3,4,5,6,7,8,9,10-decahydro-10-(4-hydroxyphenyl)-3,3-dimethyl-1,8-dioxoacridin-9-yl)phenyl)-3,3-dimethylacridine-1,8(2H,5H,9H,10H)-dione (4i). This compound was obtained as pale yellow solid (95% ethanol), m.p. > 300°C; IR (potassium bromide, cm^{-1}): 1635 (C=O); ^{1}H NMR: δ 9.96 (s, 2H, 2OH), 7.26–7.24 (m, 2H, Ar-H), 7.13–7.10 (m, 6H, Ar-H), 6.92 (s, 4H, Ar-H), 5.03 (s, 2H, 2CH), 2.28–2.12 (m, 10H, 5CH$_2$), 2.03–1.97 (m, 4H, 2CH$_2$), 1.86–1.79 (m, 4H, 2CH$_2$), 1.69–1.60 (m, 2H, CH$_2$), 0.88 (s, 6H, 2CH$_3$), 0.70 (s, 6H, 2CH$_3$). Anal. Calcd. for C$_{48}$H$_{48}$N$_2$O$_6$: C, 76.98; H, 6.46; N, 3.74. Found: C, 77.05; H, 6.43; N, 3.75.

10-p-Tolyl-3,4,6,7-tetrahydro-9-(3-(1,2,3,4,5,6,7,8,9,10-decahydro-3,3-dimethyl-1,8-dioxo-10-p-tolylacridin-9-yl)phenyl)-3,3-dimethyl-acridine-1,8(2H,5H,9H,10H)-dione (4j). This compound was obtained as pale yellow solid (95% ethanol), m.p. > 300°C; IR (potassium bromide, cm^{-1}): 1645 (C=O); ^{1}H NMR: δ 7.38–7.34 (m, 9H, Ar-H), 7.11–7.07 (m, 1H, Ar-H), 7.02–6.98 (m, 2H, Ar-H), 5.09 (s, 2H, 2CH), 2.41 (s, 6H,

2CH$_3$), 2.21–2.14 (m, 8H, 4CH$_2$), 2.09 (s, 2H, CH$_2$), 2.01–1.94 (m, 4H, 2CH$_2$), 1.85–1.78 (m, 4H, 2CH$_2$), 1.65–1.55 (m, 2H, CH$_2$), 0.88 (s, 6H, 2CH$_3$), 0.70 (s, 6H, 2CH$_3$). Anal. Calcd. for C$_{50}$H$_{52}$N$_2$O$_4$: C, 80.61; H, 7.04; N, 3.76. Found: C, 82.49; H, 7.07; N, 3.77.

10-(4-Hydroxyphenyl)-3,4,6,7-tetrahydro-9-(3-(1,2,3,4,5,6,7,8,9,10-decahydro-10-(4-hydroxyphenyl)-3,3-dimethyl-1,8-dioxoacridin-9-yl)phenyl)-3,3-dimethylacridine-1,8(2H,5H,9H,10H)-dione (4k). This compound was obtained as pale yellow solid (95% ethanol), m.p. > 300°C; IR (potassium bromide, cm^{-1}): 1634 (C=O); ^{1}H NMR: δ 9.94 (s, 2H, 2OH), 7.34–7.23 (m, 5H, Ar-H), 7.10–6.96 (m, 3H, Ar-H), 6.91–6.89 (m, 4H, Ar-H), 5.09 (s, 2H, 2CH), 2.33–2.14 (m, 10H, 5CH$_2$), 2.05–1.98 (m, 4H, 2CH$_2$), 1.87–1.83 (m, 4H, 2CH$_2$), 1.63–1.61 (m, 2H, CH$_2$), 0.89 (s, 6H, 2CH$_3$), 0.72 (s, 6H, 2CH$_3$). Anal. Calcd. for C$_{48}$H$_{48}$N$_2$O$_6$: C, 76.98; H, 6.46; N, 3.74. Found: C, 77.03; H, 6.47; N, 3.77.

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[14] The single-crystal growth was carried out in ethanol at room temperature. X-ray crystallographic analysis was performed with a Siemens SMART CCD and a Semens P4 diffractometer. Crystal data for **4f**: $C_{46}H_{44}N_2O_6$, $M = 720.83$, Triclinic, space group P-1, $a = 11.6215(14)$, $b = 13.3902(16)$, $c = 14.7823(19)$, $V = 2171.6(5)$ Å^3, $Z = 2$, $T = 298(2)$ K, $\mu = 0.073$ mm^{-1}, 10,857 reflections measured, 7481 unique reflections, $R = 0.0981$, $R_w = 0.1263$.