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Synthesis and Transformations of New 1,2,3,4,5,6,7,8,9,10-Decahydroacridine-1,8-dione Derivatives

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Abstract—Reactions of cyclohexane-1,3-diones with amines and aldehydes led to the formation of 1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-diones containing alkylaryl, aryl, and hydroxy substituents in position *10*. Transformations of 10-hydroxy-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-diones under oxidation with nitrous acid, reduction with zinc powder, as well as on heating in aprotic solvents and acetic, phosphoric, and polyphosphoric acids, were studied. NMR spectra of 1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-diones having different substituents in positions *9* and *10* were analyzed.

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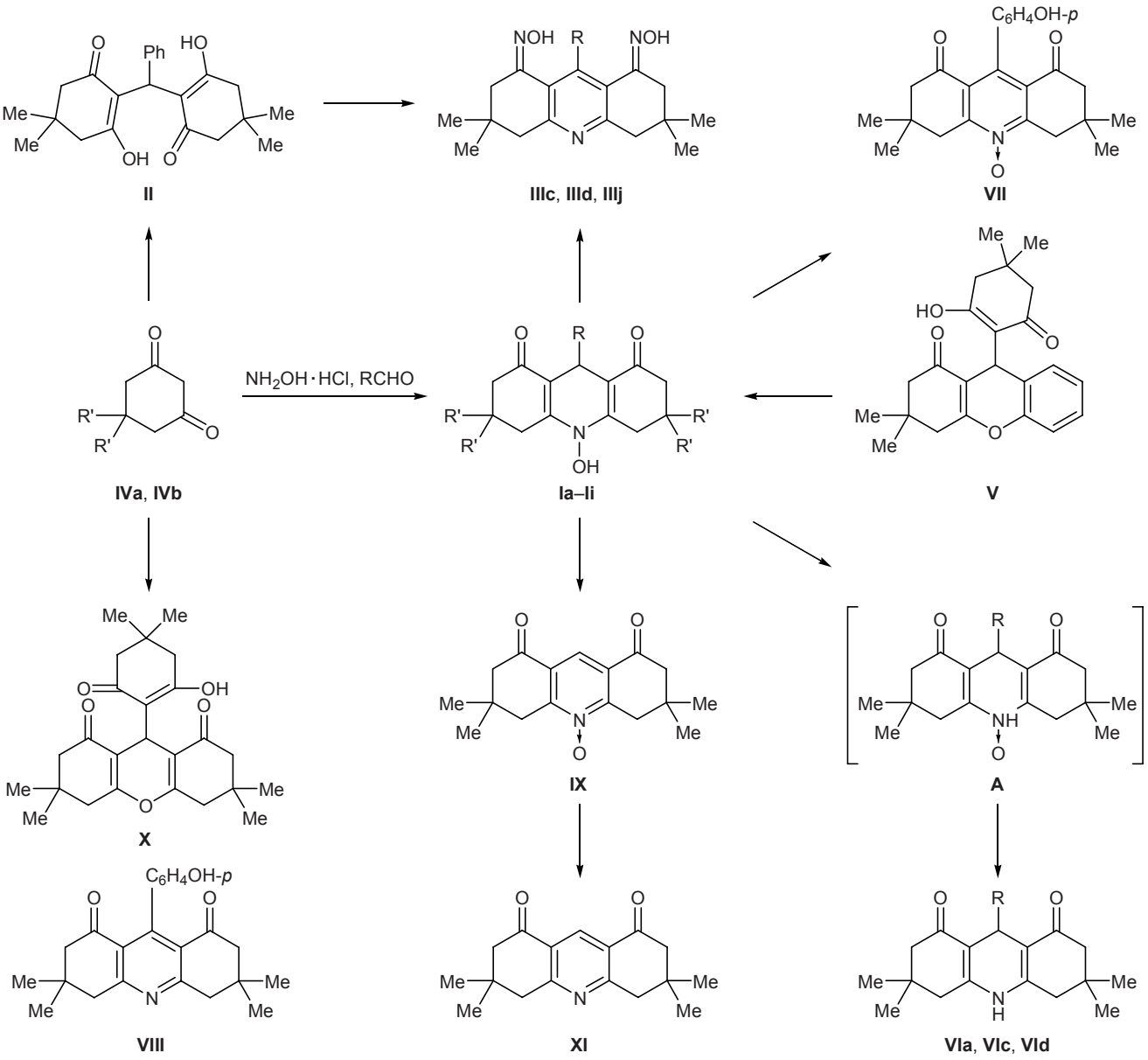
Decahydroacridinediones contain a 1,4-dihydropyridine ring as structural fragment and are available via various versions of Hantzsch synthesis [1–4]. These compounds exhibit a broad spectrum of biological activity [5, 6]. With the goal of obtaining new 10-hydroxydecahydroacridinedione derivatives **Ia**–**Ii**, in the present work hydroxylamine was used as one of the components for building up 1,4-dihydropyridine ring according to the above methods. Excess hydroxylamine hydrochloride reacted with 2,2′-phenylmethylenebis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one) (**II**) in anhydrous pyridine, as well as with the corresponding anhydride [7], to give octahydroacridinedione dioxime **IIIc** (Scheme 1). The use in this reaction of an equivalent amount of hydroxylamine resulted in the formation of 10-hydroxydecahydroacridinedione **Ic**. The most efficient procedure for the preparation of 10-hydroxydecahydroacridinediones **Ia**–**Ii** proved to be three-component condensation of cyclohexane-1,3-dione (**IVa**) or dimedone (**IVb**) with 1.2 equiv of hydroxylamine and 1.1 equiv of appropriate aldehyde in anhydrous pyridine. Initially, diketones **IVa** and **IVb** were heated with hydroxylamine in pyridine to obtain the corresponding enamino ketone, aldehyde was then added to the reaction mixture, and the mixture was heated further. Decahydroacridinedione **Ig** was also prepared by reaction of tetrahydroxanthene **V** with hydroxylamine. Heating of compound **Id** with excess hydroxylamine in pyridine gave dioxime **IIId**.

On the basis of published data [8, 9] it was expected that compounds **Ia**–**Ii** could easily lose water molecule to be converted into the corresponding octahydroacridinediones. However, they turned out to be stable (except for compound **Ie**) on heating for 3 h in acetic acid or propan-2-ol. When compounds **Ia** and **Id** were heated in aprotic solvents, oxygen elimination occurred with formation of decahydroacridinediones **VIa** and **VId**. Presumably, the reaction involved intermediate formation of N-oxide **A** [8, 9]. The reduction of *N*-hydroxyhydroacridine **Ic** with zinc powder resulted in the formation of the corresponding deoxy derivative **VIc**. The oxidation of **If** with nitrous acid according to the procedure reported in [1] gave octahydroacridinedione *N*-oxide **VII** and a minor amount of octahydroacridinedione **VIII**. Octahydroacridinedione *N*-oxide **IX** was obtained by heating compounds **Ic** and **If** in phosphoric or polyphosphoric acid as described in [10] or compound **If** in acetic acid [11]. In addition, compound **IX** was obtained from decahydroxanthene **X** (which is available from dimedone and triethyl orthoformate [12]) and hydroxylamine on heating in acetic acid. The reaction of **X** with excess hydroxylamine gave dioxime **IIIj**. The reduction of *N*-oxide **IX** with zinc powder afforded octahydroacridinedione **XI** which was identical to a sample prepared previously [10]. The structure of the obtained compounds was determined on the basis of their UV, IR, and NMR spectra and elemental analyses. *N*-Hydroxy-

Scheme 1.



R = Me (**a**), *i*-Bu (**b**), Ph (**c**, **i**), 4-$MeOC_6H_4$ (**d**), 4-$Me_2NC_6H_4$ (**e**), 4-HOC_6H_4 (**f**), 2-HOC_6H_4 (**g**), 2-$MeOC_6H_4$ (**h**), H (**j**); **Ii**, **IVa**, R′ = H; **Ia–Ih**, **IVb**, R′ = Me.

decahydroacridinediones **Ia–Ii** displayed in the UV spectra two absorption bands at λ 251–255 and 270–271 nm, which are typical of enamino ketone moiety [13]; in addition, a long-wave absorption band was observed at λ 384–406 nm, which is characteristic of 3,5-diacyl-1,4-dihydropyridine system. Addition of alkali induces red shift of the absorption maxima. The long-wave maximum shifts to λ 483–509 nm, and the solution turns purple; therefore, these compounds can be used as acid–base indicators. The magnitude of the red shift reaches 99–113 nm; the corresponding shift for unsubstituted derivatives **VIc**, **VId**, and **XIIg** amounts to 78–83 nm (see table), while the intensity of the initial band remains high at the same alkali concentration (10^{-2} M), indicating high concentration of the initial form of compounds **VI**. This means that the acidity of **I** is higher than the acidity of **VI**. No shift of the absorption maxima was observed in the UV spectra of *N*-alkyl- and *N*-aryldecahydroacridinediones **XII** (see below) upon addition of alkali. A conclusion can be drawn that the observed changes in the electronic absorption spectra result from formation of anions

UV spectra of compounds **Ia–Ii, VIc, VId**, and **XIIg** in neutral and alkaline media

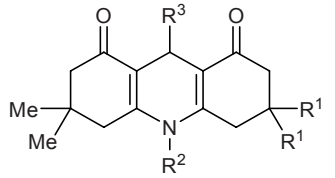
Compound no.	UV spectrum, nm (log ε)	
	EtOH	10^{-2} M KOH in EtOH
Ia	251 (4.09), 271 (3.70), 392 (3.75)	271 (4.25), 307 (3.69), 504 (3.94)
Ib	253 (4.32), 270 (3.87), 385 (3.92)	272 (4.39), 305 (3.79), 492 (4.05)
Ic	251 (4.15), 392 (3.78)	276 (4.23), 501 (3.89)
Id	226 (4.06), 251 (4.13), 270 (3.94), 392 (3.88)	225 (4.17), 270 (4.1), 301 (3.91), 502 (4.00)
Ie	251 (4.47), 390 (3.85)	258 (4.35), 302 (3.92), 502 (3.93)
If	226 (3.98), 250 (4.04), 270 (3.83), 391 (3.74)	245 (4.27), 271 (4.26), 300 (3.85), 502 (3.92)
Ig	255 (4.18), 395 (3.79)	274 (4.25), 504 (3.88)
Ih	254 (4.13), 406 (3.75)	273 (4.17), 302 (3.82), 516 (3.84)
Ii	251 (4.11), 384 (3.71)	269 (4.16), 483 (3.82)
VIc	250 (4.16), 375 (3.88)	250 (4.22), 376 (3.83), 459 (3.61)
VId	227 (4.14), 249 (4.20), 375 (3.96)	225 (4.19), 250 (4.16), 375 (3.85), 458 (3.35)
XIIg	250 (4.23), 377 (3.94)	265 (4.24), 377 (3.52), 455 (4.03)

XIII and **XIV** in alkaline solution; the presence of a negative charge in the donor part of the molecule (nitrogen atom) increases the energy of the highest occupied molecular orbital (HOMO), which is responsible for the red shift of the charge-transfer band [14].

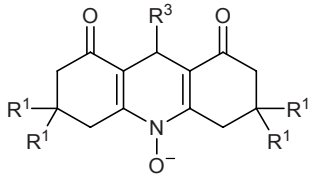
In the IR spectra of decahydroacridinediones **Ia–Ii**, absorption bands in the region 1620–1670 cm^{-1} correspond to the enamino ketone fragments. The carbonyl absorption band in the IR spectra of octahydroacridinediones is located at 1700–1725 cm^{-1}; such a frequency is fairly high for a carbonyl group conjugated with aromatic ring. Presumably, interaction between two identical carbonyl functions (resonance effect) leads to band splitting [15].

The presence of a 3,5-diacyl-1,4-dihydropyridine fragment in molecules **Ia–Ii** is responsible for luminescence of these compounds. Like their *N*-phenyl-substituted analogs [16], luminescence of **Ia–Ii** is observed under UV irradiation. For example, irradiation of a solution of **Id** in alcohol (λ_{max} 373 nm) gives rise to luminescence with its maximum at λ_{fl} 464 nm. Irradiation of the same compound in a 10^{-2} N solution of sodium hydroxide (λ_{max} 502 nm) induces fluorescence at λ_{fl} 696 nm.

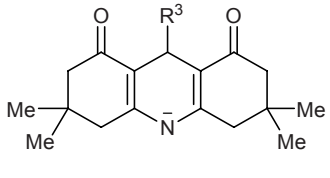
The NMR spectra of decahydroacridinediones **Ia–Ii** correspond to structures with a symmetry plane passing through the C^9 and N^{10} atoms. On the other hand, the NMR spectra of octahydroacridine derivatives indicate that their molecules have one more symmetry plane passing through all 14 atoms of the acridine skeleton and orthogonal to the first symmetry plane; therefore, their structure is characterized by the second-order symmetry axis, and it corresponds to the C_{2v} point group symmetry [17]. In the ^1H NMR spectra of **Ia** and **Ic–Ih**, four methyl groups in positions *3* and *6* appear as two singlets, while in the spectra of **III** and **VII**–**IX**, protons in the methyl groups give only one singlet. At least one signal from two equivalent methylene groups (C^2H_2/C^8H_2 and C^4H_2/C^5H_2) in compounds **Ia–Ih** appears as an *AB* quartet. Compounds **III** and **VII–IX** display two singlets from the methylene protons. Obviously, nonequivalence of geminal



XIIa–XIIi



XIIIa–XIIIi



XIVa, **XIVc**, **XIVd**

XII, R^1 = H (**c**), Me (**a**, **b**, **d**–**i**); R^2 = $PhCH_2$ (**a**, **c**, **d**), 3,4-$(MeO)_2C_6H_3CH_2CH_2$ (**b**), $HOCH_2CH_2$ (**e**), Ph (**f**), H (**g**), $PhCH_2CH_2$ (**h**, **i**); R^3 = H (**a**), 3,4-$(MeO)_2C_6H_3$ (**b**, **h**), *i*-Pr (**c**, **d**), 2-HOC_6H_4 (**e**–**g**, **i**); **XIII**, **XIV**, R^1 = H (**i**), Me (**a**–**h**); R^3 = Me (**a**), *i*-Bu (**b**), Ph (**c**, **i**), 4-$MeOC_6H_4$ (**d**), 4-$Me_2NC_6H_4$ (**e**), 4-HOC_6H_4 (**f**), 2-HOC_6H_4 (**g**), 2-$MeOC_6H_4$ (**h**).

protons and methyl groups in decahydroacridinediones **Ia**–**Ii** originates from the effect of different substituents in position *9*. Inversion of the rings does not give two equivalent structures, and the plane of the acridine skeleton does not become a symmetry plane.

Analysis of published data shows that our spectral data are not always consistent with the data reported by other authors for the same structures. In particular, Shchekotikhin and Nikolaeva [7] reported the ^{1}H NMR spectrum of octahydroacridinedione **IIIc**, which contained two singlets from four methyl groups in positions *3* and *6*; compound **IIIc** isolated in the present work showed only one singlet in the same spectral region. In the spectra of decahydroacridinedione derivatives **XII** having an *ortho*-substituted phenyl ring, more than two signals from methyl groups (3 to 4) were observed [4]. The compounds having no substituent on C^9 and an *ortho*-substituted phenyl group on the nitrogen atom revealed nonequivalence of the C^2H_2/C^8H_2 and C^4H_2/C^5H_2 methylene groups [18] and geminal methyl groups on C^3 and C^6 [2, 6]. This may be due to the presence of different rotamers arising from restricted rotation of the aromatic substituent about the C–N bond [4]. The ^{1}H NMR spectrum of 10-benzyl-substituted decahydroacridinedione **XIIa** reported in [2] contained two singlets from the four methyl groups, though only one singlet should be expected on the basis of the above considerations, taking into account the lack of substituent in position *9*.

We synthesized compound **XIIa** and a series of its *N*-aralkyl-substituted analogs **XIIb–XIIi** with a view to examine their NMR spectra. The syntheses were performed by three-component condensation of dimedone or cyclohexane-1,3-dione, appropriate 3-aminocyclohex-2-en-1-one, and aldehyde in acetic acid. Compound **XIIi** was obtained by reaction of **V** with 2-phenylethanamine [19]. In the ^{1}H NMR spectrum of decahydroacridinedione **XIIa** only one singlet from methyl protons was observed. The spectra of compounds **XIIb–XIIi** indicated that their molecules have one symmetry plane with no rotamers. The same applies for their *N*-alkyl derivatives [19]. The ^{1}H and ^{13}C NMR spectra of **XIIc** possessing a chiral center (C^9) contained two signals from the methyl groups in the isopropyl substituent on C^9, while the corresponding groups in symmetric analog **XIId** gave rise to a single peak. Octahydroxanthene derivatives having an oxygen atom instead of nitrogen in position *10* displayed similar spectral patterns [19].

EXPERIMENTAL

The UV spectra were recorded on a Specord M-400 spectrophotometer. The fluorescence spectra were measured on a Solar instrument. The IR spectra were obtained in KBr on a UR-20 spectrometer. The ^{1}H and ^{13}C NMR spectra of compounds **XIIb–XIId** were recorded on a Bruker Avance 500 spectrometer at 500 and 125 MHz, respectively, and the other compounds were examined on a Bruker AC-200 spectrometer at 200 and 50 MHz, respectively; tetramethylsilane was used as internal reference. The progress of reactions and the purity of products were monitored by TLC on Silufol UV-254 plates using EtOAc–hexane (1:1) as eluent; spots were visualized under UV radiation or by treatment with iodine vapor, followed by calcination at 250–350°C. The melting points were determined on a Boetius hot stage.

10-Hydroxy-3,3,6,6-tetramethyl-9-phenyl-1,2,-3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (Ic). *a*. A mixture of 1.10 g (3 mmol) of tetraketone **II** and 0.21 g (3 mmol) of hydroxylamine hydrochloride in 20 ml of pyridine was heated for 2 h under reflux. The mixture was cooled to room temperature, diluted with 150 ml of water, and left to stand for 24 h. The precipitate was filtered off, washed with water (150 ml), dried in air, and recrystallized from ethanol. Yield 0.68 g (62%), mp 254–256ºC. IR spectrum, ν, cm^{-1}: 1150, 1235, 1380, 1620–1660 (C=C, C=O), 3250 (OH). ^{1}H NMR spectrum (CD_3CO_2D), δ, ppm: 0.94 s (6H, Me), 1.10 s (6H, Me), 2.21 and 2.33 (2H each, CH_2, *AB* system, J_{AB} = 17 Hz), 2.56 and 2.84 (2H each, CH_2, *AB* system, J_{AB} = 17 Hz), 5.14 s (1H, 9-H), 7.00–7.20 (5H, H_{apom}). ^{13}C NMR spectrum (CD_3CO_2D), δ_C, ppm: 27.01 (Me), 29.96 (Me), 32.79 (C^3, C^6), 33.62 (C^9), 38.52 (C^4, C^5), 50.05 (C^2, C^7), 112.44 (C^{8a}, C^{9a}), 127.29 ($C^{4'}$), 128.77 (C_{apom}), 129.01 (C_{apom}), 146.34 ($C^{1'}$), 155.50 (C^{4a}, C^{10a}), 199.01 (C^1, C^8). Found, %: C 75.72; H 7.34; N 3.80. $C_{23}H_{27}NO_3$. Calculated, %: C 75.59; H 7.45; N 3.83.

b. A mixture of 2.8 g (20 mmol) of 5,5-dimethylcyclohexane-1,3-dione (dimedone) and 0.76 g (11 mmol) of hydroxylamine hydrochloride in 15 ml of pyridine was heated for 1 h under reflux, 1.17 g (11 mmol) of benzaldehyde was added, and the mixture was heated for 2 h more under reflux. It was then cooled to room temperature, diluted with 150 ml of water, and left to stand for 24 h. The precipitate was filtered off, washed with water (150 ml), and dried in air. Yield 3.32 g (91%).

10-Hydroxy-3,3,6,6,9-pentamethyl-9-phenyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (Ia) was synthesized in a similar way from 2.80 g of dimedone, 0.83 g of hydroxylamine hydrochloride, and 0.48 g of acetaldehyde. Yield 2.58 g (85%), mp 223–225°C. IR spectrum, ν, cm^{-1}: 1150, 1240, 1380, 1620–1660 (C=C, C=O), 2900, 3200 (OH). ^1H NMR spectrum (CD_3CO_2D), δ, ppm: 0.92 d (3H, 9-Me, J = 7 Hz), 1.04 s (6H, Me), 1.07 s (6H, Me), 2.06 s (4H, CH_2), 2.56 and 2.72 (2H each, CH_2, *AB* system, J_{AB} = 17 Hz), 4.00 q (1H, 9-H, J = 7 Hz). ^{13}C NMR spectrum (CD_3CO_2D), δ_C, ppm: 22.72 (9-Me), 26.86 (Me), 29.86 (Me), 29.96 (C^9), 32.83 (C^3, C^6), 38.46 (C^4, C^5), 50.15 (C^2, C^7), 113.36 (C^{8a}, C^{9a}), 156.04 (C^{4a}, C^{10a}), 199.21 (C^1, C^8). Found, %: C 67.41; H 8.58; N 4.25. $C_{18}H_{25}NO_3 \cdot H_2O$. Calculated, %: C 67.26; H 8.47; N 4.36.

10-Hydroxy-3,3,6,6-tetramethyl-9-(2-methylpropyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (Ib) was obtained in a similar way from 2.80 g of dimedone, 0.83 g of hydroxylamine hydrochloride, and 0.95 g of 3-methylbutanal. Yield 3.00 g (87%), mp 242–244°C. IR spectrum, ν, cm^{-1}: 1150, 1235, 1385, 1605 (C=C), 1660 (C=O), 3250 (OH). ^1H NMR spectrum (CF_3CO_2D–CD_3OD, 1:1), δ, ppm: 0.90 d (6H, Me, J = 6.5 Hz), 1.16 s (12H, Me), 1.20 m (2H, 1′-H), 1.42 m (2′-H), 2.34 s (4H, CH_2), 2.62 and 2.80 (2H each, CH_2, *AB* system, J_{AB} = 18 Hz) 4.15 t (1H, 9-H, J = 6.5 Hz). ^{13}C NMR spectrum (CF_3CO_2D–CD_3OD, 1:1), δ_C, ppm: 23.63 (2′-Me), 25.38 (C), 25.81 (C), 27.18 (Me), 30.26 (Me), 33.21 (C^3, C^6), 39.17 (C^4, C^5), 47.29 (9-CH_2), 51.02 (C^2, C^7), 113.43 (C^{8a}, C^{9a}), 157.52 (C^{4a}, C^{10a}), 198.90 (C^1, C^8). Found, %: C 69.33; H 9.00; N 3.82. $C_{21}H_{31}NO_3 \cdot H_2O$. Calculated, %: C 69.39; H 9.15; N 3.85.

10-Hydroxy-9-(4-methoxyphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (Id) was synthesized in a similar way from 2.80 g of dimedone, 0.83 g of hydroxylamine hydrochloride, and 1.50 g of 4-methoxybenzaldehyde. Yield 3.52 g (89%), mp 228–230°C. IR spectrum, ν, cm^{-1}: 1150, 1230, 1375, 1615–1660 (C=C, C=O), 3250 (OH). ^1H NMR spectrum (CF_3CO_2D–CD_3OD, 1:1), δ, ppm: 1.02 s (6H, Me), 1.18 s (6H, Me), 2.44 and 2.28 (2H each, CH_2, *AB* system, J_{AB} = 17 Hz), 2.66 and 2.98 (2H each, CH_2, *AB* system, J_{AB} = 17 Hz), 3.74 s (3H, OMe), 5.14 s (1H, 9-H), 6.79 d (2H, H_{arom}, J = 8 Hz), 7.22 d (2H, H_{arom}, J = 8 Hz). ^{13}C NMR spectrum (CF_3CO_2D–CD_3OD, 1:1), δ_C, ppm: 27.49 (Me), 29.72 (Me), 35.58 (C^3, C^6, C^9), 39.19 (C^4, C^5), 50.13 (C^2, C^7), 56.12 (OMe), 113.71 (C^{8a}, C^{9a}), 115.30 (C_{arom}), 119.75 (C_{arom}), 130.51 (C_{arom}), 158.43 (C^p), 161.70 (C^{4a}, C^{10a}), 199.31 (C^1, C^8). Found, %: C 72.76; H 7.29; N 3.43. $C_{24}H_{29}NO_4$. Calculated, %: C 72.88; H 7.39; N 3.54.

10-Hydroxy-9-(4-dimethylaminophenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (Ie) was synthesized in a similar way from 2.80 g of dimedone, 0.83 g of hydroxylamine hydrochloride, and 1.64 g of 4-dimethylaminobenzaldehyde. Yield 3.67 g (90%), mp 165–167°C. IR spectrum, ν, cm^{-1}: 1150, 1235, 1375, 1620–1660, 3250, 3300. ^1H NMR spectrum (CD_3OD), δ, ppm: 0.96 s (6H, Me), 1.12 s (6H, Me), 2.11 and 2.27 (2H each, CH_2, *AB* system, J_{AB} = 17 Hz), 2.67 and 2.79 (2H each, CH_2, *AB* system, J_{AB} = 17 Hz), 3.25 s (6H, NMe_2), 5.16 s (1H, 9-H), 7.54 m (4H, H_{arom}). Found, %: C 73.37; H 7.82; N 6.94. $C_{25}H_{32}N_2O_3$. Calculated, %: C 73.49; H 7.90; N 6.86.

10-Hydroxy-9-(4-hydroxyphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (If) was synthesized in a similar way from 2.80 g of dimedone, 0.83 g of hydroxylamine hydrochloride, and 1.34 g of 4-hydroxybenzaldehyde. Yield 3.51 g (92%), mp 244–246°C. IR spectrum, ν, cm^{-1}: 1230, 1370, 1520, 1550, 1620 (C=C), 1670 (C=O), 3200 (OH). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 0.90 s (6H, Me), 1.04 s (6H, Me), 2.01 and 2.19 (2H each, CH_2, *AB* system, J_{AB} = 17 Hz), 2.55 and 2.66 (2H each, CH_2, *AB* system, J_{AB} = 18 Hz), 4.90 s (1H, 9-H), 6.58 d (2H, H_{arom}, J = 8 Hz), 7.02 d (2H, H_{arom}, J = 8 Hz), 9.18 br.s (OH), 10.96 br.s (OH). ^{13}C NMR spectrum (DMSO-d_6), δ_C, ppm: 26.65 (Me), 29.36 (Me), 30.62 (C^9), 31.56 (C^3, C^6), 39.01 (C^4, C^5), 49.63 (C^2, C^7), 110.00 (C^{8a}, C^{9a}), 114.44 (C_{arom}), 128.06 (C_{arom}), 136.51 (C^i), 152.09 (C^p), 155.16 (C^{4a}, C^{10a}), 196.00 (C^1, C^8). Found, %: C 73.68; H 6.27; N 4.61. $C_{19}H_{19}NO_3$. Calculated, %: C 73.76; H 6.19; N 4.53.

10-Hydroxy-9-(2-hydroxyphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (Ig). *a*. Compound **Ig** was synthesized as described above for **Ic** from 2.80 g of dimedone, 0.83 g of hydroxylamine hydrochloride, and 1.34 g of salicylaldehyde. Yield 3.47 g (91%), mp 203–205°C. IR spectrum, ν, cm^{-1}: 1150, 1240, 1380, 1495, 1600, 1630–1660 (C=C, C=O), 3260 (OH). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 0.97 s (6H, Me), 1.11 s (6H, Me), 2.05 and 2.22 (2H each, CH_2, *AB* system, J_{AB} = 17 Hz), 2.45 and 2.70 (2H each, CH_2, *AB* system, J_{AB} = 18 Hz), 5.08 s (1H, 9-H), 6.65 m (1H, H_{arom}), 6.90 m (2H, H_{arom}), 7.29 m (1H, H_{arom}). ^{13}C NMR spectrum

(DMSO-d_6), δ_C, ppm: 26.91 (Me), 29.46 (Me), 29.94 (C^9), 32.64 (C^3, C^6) 35.98 (C^4, C^5), 50.35 (C^2, C^7), 108.34 (C^{8a}, C^{9a}), 109.31 (C_{arom}), 111.83 (C_{arom}), 120.39 C_{arom}), 128.26 (C_{arom}), 137.07 (C_{arom}), 153.32 (COH), 154.03 (C^{4a}, ^{10a}), 196.91 (C^1, C^8). Found, %: C 72.30; H 7.05; N 3.77. $C_{23}H_{27}NO_4$. Calculated, %: C 72.42; H 7.13; N 3.67.

b. A mixture of 0.73 g (2 mmol) of compound **V** and 0.15 g (2.2 mmol) of hydroxylamine hydrochloride in 20 ml of pyridine was heated for 4 h under reflux. The mixture was then cooled to room temperature, diluted with 150 ml of water, and left to stand for 24 h. The precipitate was filtered off, washed with water (150 ml), dried in air, and recrystallized from ethanol. Yield 0.57 g (75%).

10-Hydroxy-9-(2-methoxyphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (Ih) was synthesized in a similar way from 2.80 g of dimedone, 0.83 g of hydroxylamine hydrochloride, and 1.50 g of 2-methoxybenzaldehyde. Yield 3.48 g (88%), mp 247–249°C. IR spectrum, ν, cm^{-1}: 1150, 1230, 1370, 1500, 1610–1670 (C=C, C=O), 3250, 3450 (OH). ^{1}H NMR spectrum (CF_3CO_2D–CD_3OD, 1:1), δ, ppm: 0.92 s (6H, Me), 1.12 s (6H, Me), 2.08 and 2.26 (2H each, CH_2, *AB* system, J_{AB} = 17 Hz), 2.56 and 2.80 (2H each, CH_2, *AB* system, J_{AB} = 18 Hz), 3.80 s (3H, OMe), 5.16 s (1H, 9-H), 6.84 m (2H, H_{arom}), 7.08 m (1H, H_{arom}), 7.38 m (1H, H_{arom}). ^{13}C NMR spectrum (CF_3CO_2D–CD_3OD, 1:1), δ_C, ppm: 26.63 (Me), 29.82 (Me), 30.26 (C^9), 33.25 (C^3, C^6), 38.91 (C^4, C^5), 50.00 (C^2, C^7), 55.72 (OMe), 111.37 (C^{8a}, C^{9a}), 111.91 (C_{arom}), 112.00 (C_{arom}), 120.93 (C_{arom}), 128.86 (C_{arom}), 133.67 (C_{arom}), 156.13 (**C**OMe), 160.15 (C^{4a}, C^{10a}), 199.10 (C^1, C^8). Found, %: C 72.75; H 7.47; N 3.62. $C_{24}H_{29}NO_4$. Calculated, %: C 72.88; H 7.39; N 3.54.

10-Hydroxy-9-phenyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (Ii) was synthesized in a similar way from 2.24 g of cyclohexane-1,3-dione, 0.83 g of hydroxylamine hydrochloride, and 1.17 g of benzaldehyde. Yield 2.66 g (86%), mp 254–256°C. IR spectrum, ν, cm^{-1}: 1140, 1185, 1240, 1310, 1370, 1620 (C=C), 1635, 1660 (C=O), 3230 (OH). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 1.78 m (2H), 1.96 m (2H), 2.43 m (4H), 2.68 m (2H), 2.83 d.t (2H, J_1 = 18, 5 Hz), 5.09 s (1H, 9-H), 7.07–7.19 (5H, H_{arom}), 10.81 br.s (1H, OH). ^{13}C NMR spectrum (DMSO-d_6), δ_C, ppm: 22.50 (CH_2), 30.41 (C^9), 36.62 (CH_2), 37.27 (CH_2), 110.42 (C^{8a}, C^{9a}), 126.36 (C_{arom}), 126.52 (C_{arom}), 127.68 (C_{arom}), 146.01 (C^i), 154.1 (C^{4a}, C^{10a}), 194.62 (C^1, C^8). Found, %: C 72.30; H 7.05; N 3.77. $C_{23}H_{27}NO_4$. Calculated, %: C 72.42; H 7.13; N 3.67.

3,3,6,6-Tetramethyl-9-phenyl-1,2,3,4,5,6,7,8-octahydroacridine-1,8-dione dioxime (IIIc). A mixture of 1.10 g (3 mmol) of tetraketone **II** and 1.00 g (14.4 mmol) of hydroxylamine hydrochloride in 20 ml of pyridine was heated for 3 h under reflux. The mixture was then cooled to room temperature, diluted with 150 ml of water, and left to stand for 24 h. The precipitate was filtered off, washed with 150 ml of water, dried in air, and recrystallized from ethanol. Yield 0.90 g (80%), mp 263–265°C (from EtOH); published data [1, 8]: mp 250°C. IR spectrum, ν, cm^{-1}: 1250, 1375, 1490, 1555, 1620 (C=C), 1650 (C=O), 2875, 2965 (CH), 3200–3400 (OH). UV spectrum (EtOH), λ_{max}, nm (logε): 251 (4.06), 310 (3.50). ^{1}H NMR spectrum (CF_3CO_2D), δ, ppm: 1.25 s (12H, Me), 3.02 s (4H, 2-H, 7-H), 3.20 s (4H, 4-H, 5-H), 7.20–7.60 m (5H, H_{arom}). ^{13}C NMR spectrum (CF_3CO_2D), δ_C, ppm: 29.76 (Me); 32.85 (C^3, C^6); 39.38 (C^2, C^7); 43.41 (C^4, C^5); 130.82, 131.31, 132.12 (C_{arom}); 129.81, 135.19 (C^9, C^{8a}, C^{9a}); 154.57 (C^{4a}, C^{10a}); 157.88 (C^1, C^8). Found, %: C 73.22; H 7.30; N 11.20. $C_{23}H_{27}N_3O_2$. Calculated, %: C 73.18; H 7.21; N 11.13.

3,3,6,6-Tetramethyl-9-(4-methoxyphenyl)-1,2,3,4,5,6,7,8-octahydroacridine-1,8-dione dioxime (IIId). A mixture of 0.79 g (2 mmol) of diketone **Id** and 0.56 g (8 mmol) of hydroxylamine hydrochloride in 20 ml of pyridine was heated for 3 h under reflux. The mixture was cooled to room temperature, diluted with 150 ml of water, and left to stand for 24 h. The precipitate was filtered off, washed with 150 ml of water, dried in air, and recrystallized from ethanol. Yield 0.66 g (81%), mp 268–270°C (from EtOH). IR spectrum, ν, cm^{-1}: 1250, 1280, 1375, 1390, 1475, 1520, 1555, 1590 (C=C), 1620 (C=O), 2875, 2965 (CH), 3200–3300 (OH). UV spectrum (EtOH), λ_{max}, nm (logε): 228 (4.34), 257 (4.24), 312 (3.78). ^{1}H NMR spectrum (CF_3CO_2D), δ, ppm: 1.16 s (12H, Me), 2.76 s (4H, 2-H, 7-H), 3.05 s (4H, 4-H, 5-H), 3.83 s (3H, OMe), 6.92 d (2H, H_{arom}, J = 8 Hz), 7.07 d (2H, H_{arom}, J = 8 Hz). ^{13}C NMR spectrum (CF_3CO_2D), δ_C, ppm: 28.65 (Me), 31.62 (C^3, C^6), 38.16 (C^2, C^7), 42.46 (C^4, C^5), 55.79 (OMe), 114.58 ($C^{3'}$, $C^{5'}$), 130.34 and 130.63 (C^{8a}, C^{9a}, $C^{1'}$), 131.73 ($C^{2'}$, $C^{6'}$), 151.75 (C^9), 154.42 (C^{4a}, C^{10a}), 156.37 (COMe), 161.04 (C^1, C^8). Found, %: C 70.67; H 7.19; N 10.23. $C_{24}H_{29}N_3O_3$. Calculated, %: C 70.73; H 7.17; N 10.31.

3,3,6,6-Tetramethyl-9-phenyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (VIc). Zinc dust,

2.10 g, was dispersed in 10 ml of 10% hydrochloric acid, and the suspension was intermittently shaken for several minutes. The liquid part was separated by decanting, and the remaining zinc material was washed with ethanol (2×10 ml). A solution of 122 mg (0.33 mmol) of compound **Ic** in 20 ml of ethanol was added to the wet residue, and the mixture was stirred for 5 h at room temperature. The precipitate was filtered off, the filtrate was evaporated under reduced pressure, and the residue was treated with water (20 ml) and chloroform (100 ml). The chloroform extract was dried with magnesium sulfate, and the solvent was distilled off. Yield 95 mg (81%), mp 290–292°C; published data [1]: mp 290–292°C.

3,3,6,6,9-Pentamethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (VIa). A mixture of 100 mg of compound **Ia** and 25 ml of dioxane was heated for 5 h under reflux. It was then cooled to room temperature and left to stand for 24 h, and the precipitate was filtered off. Yield 81 mg (85%), mp 265–268°C; published data [20]: mp 265–267°C.

9-(4-Methoxyphenyl)-3,3,6,6-tetramethyl-1,2,-3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (VId). *a*. A mixture of 100 mg of compound **Id** and 30 ml of toluene was heated for 2 h under reflux. The solution was evaporated by half and was left to stand for 24 h at room temperature, and the precipitate was filtered off. Yield 85 mg (89%), mp 269–271°C (from toluene); published data [21]: mp 270–272°C.

b. A mixture of 100 mg of compound **Id** and 15 ml of diethylene glycol dimethyl ether was heated for 30 min under reflux, the solution was diluted with 50 ml water and was left to stand for 24 h at room temperature, and the precipitate was filtered off. Yield 82 mg (85%).

Oxidation of 10-hydroxy-9-(4-hydroxyphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (If). Compound **If**, 0.5 g, was dissolved in a mixture of 10 ml of acetic and 1 ml of concentrated hydrochloric acid, the solution was cooled to 0°C, two drops of a saturated aqueous solution of sodium nitrite was added, and the mixture was kept at 0°C for 2 h and evaporated under reduced pressure, The residue was treated with 40 ml of water and 200 ml of ethyl acetate. The ethyl acetate solution was dried over magnesium sulfate and evaporated to a volume of 7 ml, and the precipitate of 9-(4-hydroxyphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroacridine-1,8-dione 10-oxide (**VII**) was filtered off. Yield 0.35 g (71%), mp 296–298°C (from EtOH). IR spectrum, ν, cm^{-1}: 1220, 1245, 1270, 1365, 1425, 1470, 1530, 1605, 1620 (C=C), 1725 (C=O), 2880, 2970 (CH), 3200–3300 (OH). UV spectrum (EtOH), λ_{max}, nm (log ε): 228 (4.30), 245 (4.38), 285 (3.88). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 1.05 s (12H, Me), 2.48 s (4H, 2-H, 7-H), 3.10 s (4H, 4-H, 5-H), 6.66 d (2H, H_{arom}, J = 8 Hz), 6.80 d (2H, H_{arom}, J = 8 Hz). Found, %: C 72.71; H 6.52; N 3.76. $C_{23}H_{25}NO_4$. Calculated, %: C 72.80; H 6.64; N 3.69.

The filtrate was subjected to column chromatography on silica gel (100–160 μm) using ethyl acetate–hexane (2:1) as eluent to isolate 0.04 g (8%) of *N*-oxide **VII** and 0.05 g (11%) of 9-(4-hydroxyphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroacridine-1,8-dione (**VIII**), mp 267–270°C (from EtOH). IR spectrum, ν, cm^{-1}: 1230, 1280, 1365, 1485, 1525, 1545, 1605, 1625 (C=C), 1720 (C=O), 2880, 2965 (CH), 3200–3300 (OH). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 1.02 s (12H, Me), 2.46 s (4H, 2-H, 7-H), 3.02 s (4H, 4-H, 5-H), 6.64 and 6.74 (2H each, CH_2, *AB* system, J_{AB} = 8 Hz), 9.32 br.s (1H, OH). Found, %: C 75.91; H 6.82; N 3.76. $C_{23}H_{25}NO_3$. Calculated, %: C 76.00; H 6.93; N 3.85.

3,3,6,6-Tetramethyl-1,2,3,4,5,6,7,8-octahydroacridine-1,8-dione 10-oxide (IX). *a*. A mixture of 1.10 g (3 mmol) of compound **Ic** and 30 g of polyphosphoric acid was stirred for 1.5 h at 115°C. It was then cooled to room temperature and cautiously diluted with 200 ml of water. After 15 h, solid impurities were filtered off, and the filtrate was neutralized with solid sodium hydroxide to pH 7–8. The precipitate was filtered off, washed with water, dried in air, and recrystallized from ethanol. Yield 0.71 g (82%), mp 223–225°C (from EtOH). IR spectrum, cm^{-1}: 1240, 1305, 1470, 1570 (C=C), 1700 (C=O), 2880, 2960 (CH). UV spectrum (EtOH), λ_{max}, nm (log ε): 224 (4.25), 249 (4.56), 296 (3.72). ^{1}H NMR spectrum (CD_2Cl_2), δ, ppm: 1.13 s (12H, Me), 2.53 s (4H, 2-H, 7-H), 3.14 s (4H, 4-H, 5-H), 8.30 s (1H, 9-H). ^{13}C NMR spectrum (CD_2Cl_2), δ_C, ppm: 28.67 (Me), 32.07 (C^3, C^6), 39.00 (C^2, C^7), 51.27 (C^4, C^5), 119.13 (C^9), 128.36 (C^{8a}, C^{9a}), 156.73 (C^{4a}, ^{10a}), 195.67 (C^1, C^8). Found, %: C 71.17; H 7.33; N 4.69. $C_{17}H_{21}NO_3$. Calculated, %: C 71.08; H 7.32; N 3.81.

b. A mixture of 0.68 g (2 mmol) of compound **If** in 28 g of 85% phosphoric acid was stirred for 1.5 h at 95°C. It was then cooled to room temperature and diluted with 200 ml of water. After 15 h, solid impurities were filtered off, the filtrate was neutralized with solid sodium hydroxide to pH 7–8, and the precipitate

was filtered off, washed with water, dried in air, and recrystallized from ethanol. Yield 0.52 g (82%).

c. A mixture of 0.41 g (1 mmol) of compound **Ie** and 10 ml of acetic acid was heated for 2 h under reflux, the solution was evaporated, and the residue was recrystallized from ethanol. Yield 0.22 g (77%).

d. A mixture of 0.82 g (2 mmol) of compound **X** [12], 0.15 g (2.2 mmol) of hydroxylamine hydrochloride, and 0.18 g (2.2 mmol) of sodium acetate in 20 ml of acetic acid was heated for 1.5 h under reflux. The solvent was distilled off, the residue was treated with 100 ml of ethanol, the precipitate of sodium chloride was filtered off, and the solution was evaporated to a volume of 7 ml. Yield 0.52 g (91%).

3,3,6,6-Tetramethyl-1,2,3,4,5,6,7,8-octahydroacridine-1,8-dione dioxime (IIIj). A mixture of 0.41 g (1 mmol) of compound **X** [12], 0.35 g (5 mmol) of hydroxylamine hydrochloride, and 0.41 g (5 mmol) of sodium acetate in 20 ml of acetic acid was heated for 2 h under reflux. The solvent was distilled off, the residue was treated with 100 ml of ethanol, the precipitate of sodium chloride was filtered off, and the filtrate was evaporated to a volume of 5 ml. Yield 0.52 g (91%), mp 278–280°C (from EtOH); published data [1]: mp 280°C. IR spectrum, ν, cm^{-1}: 1270, 1285, 1595 (C=C), 1640 (C=O), 2875, 2960 (CH), 3200–3300 (OH). UV spectrum (EtOH), λ_{max}, nm (log ε): 253 (4.22), 317 (3.63). ^{1}H NMR spectrum (CF_3CO_2D), δ, ppm: 1.22 s (12H, Me), 2.97 s (4H, 2-H, 7-H), 3.16 s (4H, 4-H, 5-H), 9.31 (1H, 9-H). ^{13}C NMR spectrum (CF_3CO_2D), δ_C, ppm: 28.47 (Me), 32.13 (C^3, C^6), 37.66 (C^2, C^7), 42.66 (C^4, C^5), 130.12 (C^{8a}, C^{9a}), 137.45 (C^9), 153.74 (C^{4a}, C^{10a}), 156.72 (C^1, C^8). Found, %: C 67.80; H 7.59; N 13.86. $C_{17}H_{23}N_3O_2$. Calculated, %: C 67.75; H 7.69; N 13.94.

3,3,6,6-Tetramethyl-1,2,3,4,5,6,7,8-octahydroacridine-1,8-dione (XI). Zinc dust, 2.10 g, was dispersed in 10 ml of 10% hydrochloric acid, and the suspension was intermittently shaken for several minutes. The liquid part was separated by decanting, and the remaining zinc material was washed with ethanol (2× 10 ml). A solution of 90 mg (0.33 mmol) of *N*-oxide **IX** in 20 ml of ethanol was added to the wet residue, and the mixture was stirred for 7 h at room temperature. The precipitate was filtered off, the filtrate was evaporated under reduced pressure, and the residue was treated with 20 ml of water and 100 ml of chloroform. The chloroform extract was dried over magnesium sulfate and evaporated. Yield 95 mg (81%), mp 145–147°C (from chloroform–hexane); published data [10]: mp 144–145°C.

10-Benzyl-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,-9,10-decahydroacridine-1,8-dione (XIIa). A mixture of 2.80 g (20 mmol) of dimedone, 1.07 g (10 mmol) of benzylamine, and 0.30 g of paraformaldehyde in 20 ml of glacial acetic acid was heated for 50 min under reflux. The solvent was distilled off, and the residue was recrystallized from ethyl acetate. Yield 1.10 g (30%), mp 198–200°C; published data [2]: mp 168–170°C. IR spectrum, ν, cm^{-1}: 1175, 1200, 1255, 1390, 1575, 1600 (C=C), 1635 (C=O), 2870, 2960 (CH). UV spectrum (MeOH), λ_{max}, nm (log ε): 247 (4.12), 278 (3.90), 390 (3.75). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 0.98 s (12H, Me), 2.26 s (4H, CH_2), 2.30 s (4H, CH_2), 3.18 s (1H, 9-H), 4.80 s (NCH_2), 7.05–7.38 (5H, H_{arom}). Found, %: C 79.36; H 7.95; N 3.80. $C_{24}H_{29}NO_2$. Calculated, %: C 79.30; H 8.04; N 3.85.

9-(3,4-Dimethoxyphenyl)-10-[2-(3,4-dimethoxyphenyl)ethyl]-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,-9,10-decahydroacridine-1,8-dione (XIIb). A mixture of 2.80 g (20 mmol) of dimedone and 1.81 g (10 mmol) of 2-(3,4-dimethoxyphenyl)ethanamine in 20 ml of gla-cial acetic acid was heated for 30 min under reflux, 1.66 g (10 mmol) of 3,4-dimethoxybenzaldehyde was added, and the mixture was heated under reflux for an additional 1.5 h. The solvent was distilled off, and the residue was recrystallized from ethyl acetate. Yield 4.07 g (71%), mp 147–149°C (from EtOAc). IR spectrum, ν, cm^{-1}: 1145, 1160, 1200, 1235, 1270, 1360, 1380, 1470, 1520, 1575 (C=C), 1635 (C=O), 2875, 2960 (CH). UV spectrum (EtOH), λ_{max}, nm (log ε): 231 (4.36), 253 (4.24), 277 (4.16), 378 (3.82). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 0.98 s (6H, Me), 1.09 s (6H, Me), 2.23 s (4H, CH_2), 2.41 and 2.48 (2H each, CH_2 *AB* system, J_{AB} = 17 Hz), 2.83 t (2H, CH_2, J = 7 Hz), 3.79 s (3H, OMe), 3.80 s (3H, OMe), 3.84 s (3H, OMe), 3.86 s (3H, OMe), 3.90 t (2H, CH_2, J = 7 Hz), 5.25 s (1H, 9-H), 6.63–6.66 m (2H, H_{arom}), 6.70–6.78 m (3H, H_{arom}), 7.00 s (1H, H_{arom}). ^{13}C NMR spectrum ($CDCl_3$), δ_C, ppm: 27.72, 29.56 (Me); 31.13 (C^9); 32.32 (C^3, C^6); 37.43 (C^4, C^5); 40.64, 46.28 (CH_2); 49.88 (C^2, C^7); 55.77, 55.83, 55.93, 55.97 (OMe); 110.71, 111.66, 111.79, 111.89 (C_{arom}); 115.40 (C^{8a}, C^{9a}); 118.89, 120.75, 129.58, 138.72, 148.30, 148.52, 149.30, 149.74; 162.19 (C^{4a}, C^{10a}); 195.70 (C^1, C^8). Found, %: C 73.33; H 7.49; N 2.51. $C_{35}H_{43}NO_6$. Calculated, %: C 73.27; H 7.56; N 2.44.

10-Benzyl-9-isopropyl-3,3-dimethyl-1,2,3,4,5,6,-7,8,9,10-decahydroacridine-1,8-dione (XIIc). A mixture of 687 mg (3 mmol) of 3-benzylamino-5,5-dimethylcyclohex-2-en-1-one, 336 mg (3 mmol) of cyclohexane-1,3-dione, and 259 mg (3.6 mmol) of

isobutyraldehyde in glacial acetic acid was heated for 1 h under reflux. The solvent was distilled off, and the residue was recrystallized from ethyl acetate. Yield 690 mg (61%), mp 112–114°C (from EtOAc). IR spectrum, ν, cm^{-1}: 1185, 1245, 1350, 1385, 1460, 1570 (C=C), 1645 (C=O), 2875, 2965 (CH). UV spectrum (EtOH), λ_{max}, nm (logε): 253 (4.24), 270 (4.12), 377 (3.79). ^1H NMR spectrum ($CDCl_3$), δ, ppm: 0.74 d (3H, Me, J = 7 Hz), 0.75 d (3H, Me, J = 7 Hz) 1.01 s (3H, Me), 1.04 s (3H, Me), 1.52 m (1H, CH), 1.90–2.72 m (10H, CH_2), 4.19 s (1H, 9-H), 4.87 s (2H, CH_2N), 6.80–7.20 m (5H, H_{arom}). ^{13}C NMR spectrum ($CDCl_3$), δ_C, ppm: 19.46 (Me); 21.52, 26.94 (CH_2); 28.23, 28.99, 30.61 (Me); 32.73 (C^3, C^6); 34.81 (C^9); 36.89, 40.35, 49.16, 50.48 (CH_2); 114.33, 114.68, 125.48 (C_{arom}); 127.88 (CH_{arom}); 129.29 (CH); 137.32 (C_{arom}); 151.56, 153.72 (C^{4a}, C^{10a}); 196.50, 196.70 (C^1, C^8). Found, %: C 79.60; H 8.19; N 3.75. $C_{25}H_{31}NO_2$. Calculated, %: C 79.53; H 8.28; N 3.71.

10-Benzyl-9-isopropyl-3,3,6,6-dimethyl-1,2,3,4,-5,6,7,8,9,10-decahydroacridine-1,8-dione (XIId) was synthesized in a similar way from 916 mg (4 mmol) of 3-benzylamino-5,5-dimethylcyclohex-2-en-1-one, 560 mg (4 mmol) of dimedone, and 415 mg (4.8 mmol) of isobutyraldehyde. Yield 1199 mg (74%), mp 198–200°C (from EtOAc). IR spectrum, ν, cm^{-1}: 1155, 1185, 1220, 1245, 1375, 1400, 1480, 1575 (C=C), 1645 (C=O), 2875, 2960 (CH). UV spectrum (EtOH), λ_{max}, nm (logε): 255 (4.40), 270 (4.11), 377 (3.86). ^1H NMR spectrum ($CDCl_3$), δ, ppm: 0.75 d (6H, Me, J = 7 Hz), 0.98 s (6H, Me), 1.03 s (6H, Me), 1.52 m (1H, CH), 2.24 d (2H, 4-H, 5-H, J = 18 Hz), 2.49 d (2H, 4-H, 5-H, J = 18 Hz), 2.20 and 2.30 (2H each, 2-H, 7-H, *AB* system, J_{AB} = 18 Hz), 4.20 d (1H, 9-H, J = 7 Hz), 4.84 s (2H, CH_2N), 7.16 d (2H, H_{arom}, J = 8 Hz), 7.34 t (1H, H_{arom}, J = 8 Hz), 7.41 t (2H, H_{arom}, J = 8 Hz). ^{13}C NMR spectrum ($CDCl_3$), δ_C, ppm: 19.44, 28.07, 28.87 (Me); 30.61 (CH); 32.40 (C^3, C^6); 34.75 (C^9); 40.21, 49.03, 50.34 (CH_2); 113.83 (C_{arom}); 125.40, 127.77, 129.11 (CH_{arom}); 137.24 (C_{arom}); 151.90 (C^{4a}, C^{10a}); 196.31 (C^1, C^8). Found, %: C 79.89; H 8.78; N 3.49. $C_{27}H_{35}NO_2$. Calculated, %: C 79.96; H 8.70; N 3.45.

10-(2-Hydroxyethyl)-9-(2-hydroxyphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (XIIe) was synthesized in a similar way from 1.83 g (10 mmol) of 3-(2-hydroxyethylamino)-5,5-dimethylcyclohex-2-en-1-one, 1.40 g (10 mmol) of dimedone, and 1.34 g (11 mmol) of salicylaldehyde. Yield 3.11 g (76%), mp 209–211°C (from EtOAc). IR spectrum, ν, cm^{-1}: 1245, 1375, 1600 (C=C), 1625 (C=O), 2875, 2960 (CH), 3440 (OH). UV spectrum (MeOH), λ_{max}, nm (logε): 255 (4.26), 312 (4.39), 384 (3.93). ^1H NMR spectrum ($CDCl_3$), δ, ppm: 0.99 s (6H, Me), 1.07 s (6H, Me), 2.25 s (4H, 2-H, 7-H), 2.40 and 2.72 (2H, CH_2, *AB* system, J_{AB} = 17 Hz), 3.46 t (2H, CH_2, J = 5 Hz), 3.60 t (2H, CH_2, J = 5 Hz), 5.20 s (1H, 9-H), 6.60–7.60 m (4H, H_{arom}), 9.54 s (OH). Found, %: C 73.28; H 7.69; N 3.51. $C_{25}H_{31}NO_4$. Calculated, %: C 73.32; H 7.63; N 3.42.

9-(2-Hydroxyphenyl)-3,3,6,6-tetramethyl-10-phenyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (XIIf) was synthesized in a similar way from 2.15 g (10 mmol) of 5,5-dimethyl-3-phenylaminocyclohex-2-en-1-one, 1.40 g (10 mmol) of dimedone, and 1.34 g (11 mmol) of salicylaldehyde. Yield 3.31 g (75%), mp 272–274°C. IR spectrum, ν, cm^{-1}: 1230, 1372, 1390, 1600, 1625 (C=C), 1655 (C=O), 2875, 2960 (CH), 3160 (OH). UV spectrum (MeOH), λ_{max}, nm (logε): 255 (4.26), 385 (3.93). ^1H NMR spectrum ($CDCl_3$), δ, ppm: 0.83 s (6H, Me), 0.95 s (6H, Me), 1.90 and 2.18 (2H each, 2-H, 7-H, *AB* system, J_{AB} = 17 Hz), 1.97 and 2.17 (2H each, 4-H, 5-H, *AB* system, J_{AB} = 17 Hz), 5.32 s (1H, 9-H), 6.76–7.76 m (9H, H_{arom}), 9.45 s (OH). Found, %: C 78.88; H 7.23; N 3.11. $C_{29}H_{31}NO_3$. Calculated, %: C 78.88; H 7.08; N 3.17.

9-(2-Hydroxyphenyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (XIIg) was synthesized in a similar way from 1.39 g (10 mmol) of 3-amino-5,5-dimethylcyclohex-2-en-1-one, 1.40 g (10 mmol) of dimedone, and 1.34 g (11 mmol) of salicylaldehyde. Yield 2.96 g (86%), mp 307–309°C (from EtOAc). IR spectrum, ν, cm^{-1}: 1230, 1375, 1487, 1600, 1625 (C=C), 1650 (C=O), 2875, 2960 (CH), 3220, 3300 (OH). ^1H NMR spectrum ($CDCl_3$), δ, ppm: 1.00 s (6H, Me), 1.00 s (6H, Me), 2.07 m (4H, 2-H, 7-H), 2.30 m (4H, 4-H, 5-H), 5.16 s (1H, 9-H), 6.70–7.10 m (3H, H_{arom}), 7.54 m (1H, H_{arom}). Found, %: C 75.63; H 7.37; N 3.91. $C_{23}H_{27}NO_3$. Calculated, %: C 75.59; H 7.45; N 3.83.

9-(3,4-Dimethoxyphenyl)-3,3,6,6-tetramethyl-10-(2-phenylethyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (XIIh) was synthesized in a similar way from 2.43 g (10 mmol) of 5,5-dimethyl-3-(2-phenylethylamino)cyclohex-2-en-1-one, 1.40 g (10 mmol) of dimedone, and 1.81 g (11 mmol) of 3,4-dimethoxybenzaldehyde. Yield 3.94 g (77%), mp 172–174°C (from EtOAc). IR spectrum, ν, cm^{-1}: 1150, 1170, 1240, 1270, 1375, 1470, 1520, 1575 (C=C), 1640 (C=O), 2870, 2960 (CH). UV spectrum (MeOH), λ_{max}, nm (logε): 254 (4.30), 273 (4.18), 379 (3.91). ^1H NMR spectrum ($CDCl_3$), δ, ppm: 0.96 s (6H, Me),

1.08 s (6H, Me), 2.22 s (4H, 2-H, 7-H), 2.37 and 2.53 (2H each, 4-H, 5-H, *AB* system, J_{AB} = 17 Hz), 2.86 t (2H, CH_2, J = 7 Hz), 3.79 s and 3.83 s (3H each, OMe), 3.90 t (2H, NCH_2, J = 7 Hz), 5.25 s (1H, 9-H), 6.68–7.40 m (8H, H_{arom}). Found, %: C 77.11; H 7.58; N 2.80. $C_{33}H_{39}NO_4$. Calculated, %: C 77.16; H 7.65; N 2.73.

9-(2-Hydroxyphenyl)-3,3,6,6-tetramethyl-10-(2-phenylethyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (XIIi). A mixture of 1.83 g (5 mmol) of compound **V**, 1.21 g (10 mmol) of 2-phenylethanamine, and 5 mg of *p*-toluenesulfonic acid in 40 ml of toluene was heated for 2 h under reflux. The solvent was distilled off, and the residue was recrystallized from ethyl acetate. Yield 1.83 g (78%), mp 216–218°C (from EtOAc). IR spectrum, ν, cm^{-1}: 1243, 1378, 1600, 1630 (C=C), 1652 (C=O), 2875, 2960 (CH), 3250 (OH). UV spectrum (MeOH), λ_{max}, nm (log ε): 254 (4.28), 272 (4.04), 385 (3.78). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 0.94 s (6H, Me), 1.08 s (6H, Me), 2.24 s (4H, 2-H, 7-H), 2.46 and 2.54 (2H each, 4-H, 5-H, *AB* system, J_{AB} = 17 Hz), 2.96 t (2H, CH_2, J = 7 Hz), 4.00 t (2H, NCH_2, J = 7 Hz), 5.18 s (1H, 9-H), 6.6–7.4 m (9H, H_{arom}). Found, %: C 79.21; H 7.60; N 2.89. $C_{31}H_{35}NO_3$. Calculated, %: C 79.28; H 7.51; N 2.98.

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