wilh cold water, and recrystallized from isopropanol. The physical characteristics of perchlorates Vd, Vg, and VIa-h are summarized in Table 1.

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OCTAHYDROPYRROLO[4,3,2-m,n]ACRIDINE DERIVATIVES. 2.* 1-ARYL-4,4,8,8-TETRAMETHYL-2,3,4,5,7,8,9,10-OCTAHYDROPYRROLO[4,3,2-m,n]ACRIDIN-10-ONES AND INTERMEDIATES IN THEIR SYNTHESIS

E. A. Bisenieks, N. V. Makarova,
Ya. R. Uldrikis, and G. Ya. Dubur

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A series of 9-aroyl-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroxanthen-1,8-diones has been prepared from arylglyoxals and dimedone in a dehydrating medium; upon heating with ammonia these compounds are converted to 1-aryl-4,4,8,8-tetramethyl-2,3,4,5,7,8,9,10-octahydropyrrolo[4,3,2-m,n]acridin-10-ones. These reactions occur via the intermediate formation of tetrahydroindole derivatives.

In the present paper we continue our investigation of a novel series of nitrogen-containing heterocycles, and have synthesized a series of 1-aryl-4,4,8,8-tetramethyl-2,3,4,5,7,8,9,10-octahydropyrrolo[4,3,2-m,n]acridin-10 ones and have examined the course of their formation reactions. We have previously established the structure of this heterocyclic system by x-ray structural analysis [1] and have reported their antioxident properties [2]. The necessary intermediates for the synthesis of pyrrolo[4,3,2-m,n]acridines VI are 9-aroyl-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroxanthen-1,3-diones (III), which are themselves prepared from arylglyoxals I and 5,5-dimethylcyclohexane-1,3-dione (dimedone) in a dehydrating medium, namely, a mixture of acetic acid and acetic anhydride [3]. A decrease in the amount of acetic anhydride leads to the formation of a mixture of diones III with aroylbis(5,5-dimethylcyclohexane-1,3-dion-2-yl)methane (II) or else inhibits the reaction of arylglyoxal with dimedone (in the case of Im), so that the only pure compound IIk could be prepared using glyoxal Ik.

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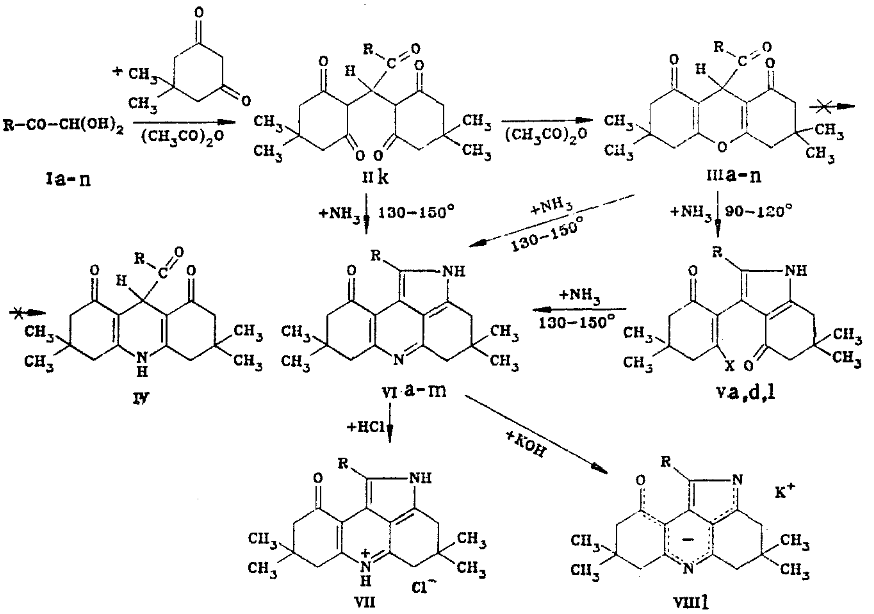

Ia, IIIa, VIa $R=C_6H_5$; b $R=2\text{-}CH_3C_6H_4$; c $R=3\text{-}CH_3C_6H_4$; d $R=4\text{-}CH_3C_6H_4$; e $R=4\text{-}CH_3OC_6H_4$; f $R=2\text{-}BrC_6H_4$; g $R=3\text{-}BrC_6H_4$; h $R=4\text{-}BrC_6H_4$; i $R=2\text{-}ClC_6H_4$; j $R=4\text{-}ClC_6H_4$; Ik—IIIk, VIk $R=2,4\text{-}Cl_2C_6H_3$; Il, IIIl, VIl, VIIIl, $R=4\text{-}NO_2C_6H_4$; Im IIIm, VIm $R=3\text{-}NO_2\text{-}4\text{-}ClC_6H_3$; In, IIIn $R=2\text{-}NO_2C_6H_4$; VII a $R=C_6H_5$, l $R=4\text{-}NO_2C_6H_4$; Va, d $X=NH_2$; Vl $X=OH$

Heating compounds II and III with ammonia in aqueous alcohol soltuion at 130-150°C leads to the formation of pyrrolo[4,3,2-m,n]acridines VI. In the formation of compounds VI the pyrrole ring is closed first. For instance, at temperatures of 90-120°C tetrahydroindole derivatives V are formed initially, and these can then be converted to pyrroloacridines VI. Closing of the 1,4-dihydropyridine ring has been detected only in the case of compound IIIn, for which formation of the pyrrole ring is sterically hindered. However, instead of the expected products IVn, Vn, or VIn, only the oxidation product could be isolated, namely, 9-(2-nitrobenzoyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroacridine-1,8-dione (IVn), in analogy with the observed formation of 9-benzoyl-10,12-dioxodiindeno[1,2-b;2',1'-e]pyridine [4].

The majority of newly synthesized pyrrolo[4,3,2-m,n]acridines VI (Tables 3-5), as with two previously studied derivatives [1], have a tendency to form variable composition crystalline solvates with ethanol and water (Table 3). In the case of the 1-phenyl derivative VIa the solvent can be removed at 80°C, whereas in the case of several other compounds in this series (VIl, m) the solvents of crystallization are removed only partially after heating for 2 h under vacuum at 120°C. For most of the compounds VI in this series, the color of the compounds is intensified and their sensitivity to photochemical decomposition increased after removal of the crystallization solvent (Table 3). Compounds in this series VI do not exhibit sharp melting points.

The UV spectra of 1-arylpyrroloacridines VIa-m (Table 5) and their 1-methyl congener [1] contain a long-wavelength absorption band in the 421-446 nm region. An excellent correlation has been observed ($n = 9$; $r = 0.997$; $\rho^* = -16.47$) between the position of the long-wavelength absorption maximum and Taft substituent constants σ^* for substituents R in m- and p-phenyl substituted derivatives VI. In selecting σ^* constants for p-substituted compounds, we used the σ^- values, which take into account polar conjugation of the substituent with the compound and which differs from the value of σ in the case of a p-nitro group [5]. This is consistent with the heightened reactivity of compound VIm with respect to anion formation in basic media. The sterically hindered derivative 1,4,4,8,8-pentamethyl-2,3,4,5,7,8,9,10-octahydropyrrolo[4,3,2-m,n]acridin-10-one [1] and all o-substituted 1-arylacridines VI do not fit this correlation.

The presence of a conjugated aminovinyliminocarbonyl system in these molecules facilitates dissociation of the NH bond of the pyrrole ring in basic media (Table 5). Anion formation proceeds more readily if there is an electron withdrawing substituent in the 1-position, and the potassium salt VIIIl can be isolated in crystalline form. This salt is stable in air, but is completely hydrolyzed in highly diluted alcohol solution. In the case of compounds VI

TABLE 1. Physical Characteristics of Compounds IIk, IIIa-n, and Va, d, *l*

Compound	mp, °C	Found, %			Molecular formula	Calculated, %			Yield, %
		C	H	N		C	H	N	
IIk	153—155	61,7	5,5		$C_{24}H_{26}Cl_2O_5$	61,9	5,6		22
IIIa	219—220	75,8	6,7		$C_{24}H_{26}O_4$	76,2	6,9		62
IIIb	176—178	76,0	7,2		$C_{25}H_{28}O_4$	76,5	7,2		16
IIIc	180—182	76,8	7,3		$C_{25}H_{28}O_4$	76,5	7,2		37
IIId	210—212	76,5	7,3		$C_{25}H_{28}O_4$	76,5	7,2		46
IIIe	154—155	73,1	6,8		$C_{25}H_{28}O_5$	73,5	6,9		48
IIIf	207—208	63,1	5,6		$C_{24}H_{25}BrO_4$	63,0	5,5		31
IIIg	172—173	63,1	5,4		$C_{24}H_{25}BrO_4$	63,0	5,5		36
IIIh	190—193	63,0	5,6		$C_{24}H_{25}BrO_4$	63,0	5,5		23
IIIi	223—224	70,1	5,8		$C_{24}H_{25}ClO_4$	69,8	6,1		16
IIIj	198—200	69,9	6,1		$C_{24}H_{25}ClO_4$	68,8	6,1		30
IIIk	203—205	64,5	5,4		$C_{24}H_{24}Cl_2O_4$	64,4	5,4		23
III*l*	196—197	68,2	6,1	3,1	$C_{24}H_{25}NO_6$	68,1	6,0	3,3	41
IIIm	192—193	62,8	5,3	2,8	$C_{24}H_{24}ClNO_6$	63,0	5,3	3,1	30
IIIn	253—255	68,7	6,1	3,2	$C_{24}H_{25}NO_6$	68,1	6,0	3,3	23
Va	214—215	73,0	7,9	7,0	$C_{24}H_{28}N_2O_2 \cdot H_2O$	73,1	7,7	7,1	40
Vd	242—244	76,5	7,8	6,9	$C_{25}H_{30}N_2O_2$	76,9	7,7	7,2	55
V*l*	207—208	67,7	6,4	6,6	$C_{24}H_{26}N_2O_5$	68,2	6,2	6,6	65

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*In the case of compounds IIk, IIIb-n, yields were calculated based on acetophenone starting material.

TABLE 2, PMR Spectra of Compounds IIIa-n in $CDCl_3$

Compound	Chemical shifts, δ, pp. (J, Hz)				
	3,6-$C(CH_3)_2$ (two s 2×6H)	4,5-CH_2 (s, 4H)	2,7-CH_2 (s 4H)	9-CH (s, 1H)	R
IIIa	1,07; 1,10	2,23	2,50	5,21	7,40—8,35 (5H, m., C_6H_5)
IIIb	1,08; 1,12	2,23	2,47	5,20	2,83 (3H, s, CH_3); 7,06—8,52 (4H, m, C_6H_4)
IIIc	1,06; 1,10	2,22	2,47	5,37	2,41 (3H, s, CH_3); 7,29—8,10 (4H, m, C_6H_4)
IIId	1,03; 1,07	2,19	2,44	5,36	2,37 (3H, s, CH_3); 7,21 (2H, d, $J=9{,}0$, 3′,5′-H); 8,11 (2H, d., $J=9{,}0$, 2′,6′-H)
IIIe	1,04; 1,08	2,19	2,44	5,35	3,83 (3H, s., OCH_3); 6,92 (2H, d, $J=9{,}0$, 3′,5′-H); 8,22 (2H, d. $J=9{,}0$, 2′,6′-H)
IIIf	1,14; 1,16	2,29	2,49	5,10	7,16—8,48 (4H, m, C_6H_4)
IIIg	1,07; 1,12	2,22	2,47	5,25	7,31—8,27 (4H, m, C_6H_4)
IIIh	1,04; 1,08	2,21	2,46	5,27	7,56 (2H, d., $J=9{,}0$, 3′,5′-H); 8,09 (2H, d., $J=9{,}0$, 2′,6′-H)
IIIi	1,09; 1,11	2,26	2,47	5,09	7,32—8,42 (4H, m, C_6H_4)
IIIj	1,05; 1,09	2,21	2,46	5,27	7,37 (2H, d., $J=9{,}0$, 3′,5′-H); 8,13 (2H, d, $J=9{,}0$, 2′,6′-H)
IIIk	1,12 (s, 12H)	2,26	2.48; 2.49 (two s 2×6H) 2×2H)	5,01	7,28—8,42 (m, C_6H_3)
III*l*	1,08; 1,13	2,20	2,47	6,21	8,32 (4H, s, C_6H_4)
IIIm	1,06; 1,11	2,22	2,47	5,16	7,63 (1H, d, $J_{56}=8{,}5$, 5′-H); 8,37 (1H, dd, $J_{65}=8{,}5$, $J_{62}=2{,}5$, 6H); 8,69 (1H, d., $J_{26}=2{,}5$, 2′-H)
IIIn	1,13; 1,18	2,30	2,50	5,17	7,40—8,44 (4H, m, C_6H_4)

containing electron donating substituents R an anhydrous medium is required for anion formation.

Compounds VI form hydrochloride salts VII (for example, hydrochloride VI*l*). In concentrated sulfuric acid partial protonation of pyrrolacridines VI is observed, as evidenced by a sharp change in their UV spectra in comparison with hydrochloride VI*l*, and the appearance of an absorption in the 530 nm region; it is difficult to study this process, however, due to decomposition of the compounds. In concentrated sulfuric acid the intermediate products III are also protonated, and their resulting UV spectra exhibit, in contrast to the UV spectra of compounds VI, pronounced bathochromic shifts, which can be as high as 70 nm in the case of the long-wavelength maximum; thus, for example, UV spectrum, λ_{max} (log ε): for IIIa (in ethanol): 204 (4.24), 244 (4.30), 293 nm (3.62); in 96% H_2SO_4: 211 (4.04), 296 (4.30), sh 310 (3.85), 355 nm (3.73); for III*l* (in ethanol): 203 (4.23), 231 (4.30), 274 nm (4.16); in 96% H_2SO_4: sh 220 (3.97), 275 (4.38), sh 345 (3.78).

TABLE 3. Physical Characteristics of Pyrrolo[4,3,2-m,n]acridines (VIa-m) and Their Salts VIIa, l and VIIIl

Compound	T_{drying}, °C	mp, °C	Compound color	Found, X			Molecular formula	Calculated, %			Yield, %
				C	H	N		C	H	N	
VIa	20	156—157	Yellow	76,6	7,9	6,6	$C_{24}H_{26}N_2O \cdot C_2H_5OH \cdot 1/2H_2O$	76,9	7,6	6,4	61
VIa	80		Orange	80,7	7,1	7,6	$C_{24}H_{26}N_2O$	80,4	7,3	7,8	
VIIa	20	Dec. >250	Orange	72,8	6,8	7,0	$C_{24}H_{26}N_2O \cdot HCl$	73,0	6,9	7,1	75
VIb	20	166—169	Yellow	76,5	8,0	6,8	$C_{25}H_{28}N_2O \cdot H_2O$	76,9	7,7	7,2	80
VIb	110		Bright yellow	79,6	7,7	7,3	$C_{25}H_{28}N_2O$	80,6	7,6	7,5	
VIc	20	255—258	Light orange	80,1	7,7	7,7	$C_{25}H_{28}N_2O$	80,6	7,6	7,5	43
VId	20	250—255	Light orange	79,9	7,9	7,3	$C_{25}H_{28}N_2O$	80,6	7,6	7,5	66
VIe	20	237—240	Yellow	71,8	8,8	6,7	$C_{25}H_{28}N_2O_2 \cdot C_2H_5OH \cdot H_2O$	71,7	8,7	6,7	59
VIe	110		Orange	76,9	7,4	7,1	$C_{25}H_{28}N_2O_2$	77,3	7,3	7,2	
VIf	20	164—167	Yellow	62,6	6,5	5,5	$C_{24}H_{25}BrN_2O \cdot C_2H_5OH \cdot 1/2H_2O$	62,9	6,8	5,4	50
VIf	120		Yellow	66,1	5,7	6,2	$C_{24}H_{25}BrN_2O$	65,9	5,8	6,4	
VIg	20	247—250	Light orange	66,9	5,9	6,1	$C_{24}H_{25}BrN_2O$	65,9	5,8	6,4	65
VIh	20	168—170	Yellow	62,5	6,6	5,5	$C_{24}H_{25}BrN_2O \cdot C_2H_5OH \cdot 1/2H_2O$	62,9	6,8	5,4	77
VIh	120		Orange	65,7	5,8	6,2	$C_{24}H_{25}BrN_2O$	65,9	5,8	6,4	
VIi	20	158—161	Yellow	70,8	6,9	6,5	$C_{24}H_{25}ClN_2O \cdot C_2H_5OH$	71,7	7,1	6,4	40
VIi	120		Yellow	73,0	6,7	7,1	$C_{24}H_{25}ClN_2O$	73,4	6,4	7,1	
VIj	20	160—165	Light orange	69,8	7,5	5,9	$C_{24}H_{25}ClN_2O \cdot 1{,}5C_2H_5OH$	70,2	7,4	6,1	74
VIj	110		Orange	72,7	6,4	6,8	$C_{24}H_{25}ClN_2O$	73,4	6,4	7,1	
VIk	20	229—232	Yellow	65,5	6,5	5,6	$C_{24}H_{24}Cl_2N_2O \cdot C_2H_5OH$	66,0	6,4	6,9	50
VIk	120		Yellow	66,4	5,5	6,2	$C_{24}H_{24}Cl_2N_2O$	67,5	5,7	6,6	
VIl	20	183—186	Orange	66,3	6,7	9,0	$C_{24}H_{25}N_3O_3 \cdot C_2H_5OH \cdot H_2O$	66,8	7,1	9,0	62
VIl	120		Dark orange	69,1	6,7	9,8	$C_{24}H_{25}N_3O_3 \cdot H_2O$	68,4	6,5	10,0	
VIIl	20	Dec. >250	Yellow	66,2	6,1	9,3	$C_{24}H_{25}N_3O_3 \cdot HCl$	65,6	6,0	9,7	86
VIIIl	20	Dec. >220	Dark brown	60,3	5,4	8,4	$C_{24}H_{24}KN_3O_3 \cdot 2H_2O$	60,1	5,9	8,8	85
VIm	20	253—255	Orange	61,5	6,0	8,6	$C_{24}H_{24}ClN_3O_3 \cdot 1/2C_2H_5OH \cdot H_2O$	62,6	6,1	8,8	63
VIm	120		Orange	64,8	5,6	9,3	$C_{24}H_{24}ClN_3O_3 \cdot 1/2H_2O$	64,5	5,6	9,4	

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*The composition and characteristics of both the solvated compounds VI, as well as samples dried in a vacuum chamber, are included in the Table.

TABLE 4. PMR Spectra of Pyrrolo[4,3,2-m,n]acridines VIa-m in $CDCl_3$*

Compound	Chemical shifts, δ, ppm (J, Hz)						
	8-CH_3 (s 6H)	4-CH_3 (s 6H)	9-CH_2 (s 2H)	5-CH_2 и 7-CH_2 (two s 2×2H)	3-CH_2 (s 2H)	2-NH br.s 1H)	R
VIa	1,09	1,14	2,45	2,83 (s, 4H)	3,03	10,00	7,26 (5H, s, C_6H_5)
VIb	1,12	1,17	2,48	2,83, 2,85	3,04	9,40	7,13—7,45 (4H, m, C_6H_4)
VIc	1,12	1,16	2,46	2,81, 2,85	3,02	9,50	2,35 (3H, s, CH_3); 6,97—7,32 (4H, m, C_6H_4)
VId	1,05	1,11	2,43	2,77, 2,79	2,99	9,76	2,32 (3H, s, CH_3); 7,08 (4H, s, C_6H_4)
VIe	1,10	1,15	2,42	2,77, 2,78	2,98	10,00	3,78 (3H, s, OCH_3); 6,82 (2H, d, J=9,0, 2′,6′-H); 7,11 (2H, d, J=9,0, 3′,5′-H)
VIf	1,09	1,17	2,38	2,80, 2,84	2,98	10,20	7,05—7,65 (4H, m, C_6H_4)
VIg	1,12	1,17	2,48	2,83, 2,85	3,04	9,40	7,13—7,45 (4H, m, C_6H_4)
VIh	1,08	1,13	2,40	2,79 (s, 4H)	2,97	10,66	6,90 (2H, d, J=9,0, 2′,6′-H); 7,20 (2H, d, J=9,0, 3′,5′-H)
VIi	1,10	1,18	2,44	2,85, 2,88	3,02	9,30	7,19—7,44 (4H, m, C_6H_4)
VIj	1,12	1,17	2,47	2,82, 2,84	3,03	9,56	7,02—7,40 (4H, m, C_6H_4)
VIk	1,13	1,20	2,43	2,82, 2,86	3,01	9,81	7,20 (2H, s, 5′,6′-H); 7,39 (1H, s, 3′-H)
VIl	1,16	1,18	2,50	2,81 (s, 4H)	3,03	10,88	7,12 (2H, d, J=9,0, 2′,6′-H); 7,91 (2H, d, J=9,0, 3′,5′-H)
VIm	1,11	1,17	2,49	2,84, 2,86	3,03	10,50	7,21 (1H, dd, J_{65}=9,0, J_{62}=2,0, 6′-H); 7,36 (1H, d, J_{56}=9,0, 5′-H); 7,71 (1H, d, J_{26}=2,0, 2′-H)

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*Chemical shifts due to solvent molecules are not reported.

TABLE 5. IR and UV Spectra of Pyrrolo[4,3,2-m,n]acridines VIa-m

Compound	IR Spectrum,* ν, cm^{-1}		UV spectrum, λ_{max},** nm (log ε)	
	NH	C=O	in ethanol	in ethanol + NaOH (0.1 M)
VIa	3400	1650	206 (4,44), 237 (4,28), 267 (4,43), 312 (3,97), 442 (3,66)	266 (4,22), 298 (4,18), 342 (3,96), 498 (3,35)
VIIa			205 (4,40), 261 (4,51), 315 (3,84), 450 (3,52)	—
VIb	3400	1663	206 (4,36), 238 (4,27), 265 (4,24), sh. 308 (3,45), 437 (3,57)	Dissociation of the NH bond is nonexistent
VIc	3405	1665	206 (4,45), 237 (4,26), 267 (4,41), sh. 308 (3,92), 442 (3,61)	Same
VId	3402	1663	206 (4,49), 238 (4,30), 268 (4,48), sh. 308 (4,05), 445 (3,65)	266 (4,32), 297 (4,29), sh 338 (4,05), 498 (3,47)
VIe	3398	1668	205 (4,50), 226 (4,26), 240 (4,26), 266 (4,49), sh. 308 (3,94), 446 (3,59)	266 (4,28), sh 290 (4,22), sh 338 (3,77), 487 (3,31)
VIf	3400	1667	206 (4,54), 238 (4,37), sh 262 (4,24), 314 (3,74), 431 (3,68)	sh 252 (4,24), 296 (4,04), 344 (3,54), 483 (3,42)
VIg	3400	1667	206 (4,52), 238 (4,29), 268 (4,33), 318 (4,05), 435 (3,65)	sh 256 (4,26), 302 (4,20), 352 (4,14), 504 (3,46)
VIh	3395	1663	205 (4,50), 226 (4,34), 236 (4,33), 269 (4,34), 319 (4,10), 437 (3,64)	sh 260 (4,21), 302 (4,19), 352 (4,13), 505 (3,42)
VIi	3400	1665	206 (4,42), 238 (4,26), 266 (4,19), 314 (3,70), 432 (3,58)	sh 254 (4,27), 287 (4,12), sh 338 (3,89), 485 (3,45)
VIj	3400	1663	206 (4,46), 224 (4,33), 238 (4,32), 269 (4,38), 317 (4,09), 439 (3,67)	sh. 258 (4,26), 302 (4,25), 349 (4.15), 505 (3,47)
VIk	3400	1663	208 (4,55), 237 (4,42), 267 (4,21), sh. 288 (3,99), 319 (3,93), 429 (3,70)	sh 246 (4,39), 303 (4,09), 355 (4,04), 490 (3,49)
VIf	3398	1663	205 (4,39), 255 (4,46), 421 (4,22)	260 (4,25), 288 (4,22), 509 (4,24)
VII1			203 (4,41), 252 (4,41), 383*** (4,03)	—
VIm	3390	1663	204 (4,38), 222 (4,38), 265 (4,30), 327 (4,07), 426 (3,69)	sh 218 (4,34), 295 (4,17), 365 (4,19), 492 (3,54)

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*In dichloroethane solution.

**Anion formation is apparently incomplete.

***Upon addition of HCl: 203 (4.47), 247 (4.39), 366 mm (4.10).

EXPERIMENTAL

IR spectra were obtained on a Perkin Elmer 580B spectrophotometer, UV spectra on a Hitachi 557 model, and PMR spectra on a Bruker WH-90 spectrometer. Mass spectra were recorded on an AEI MC-50 spectrometer.

Arylglyoxals Ib-n were synthesized according to [6] and used without further purification.

2,4-Dichlorobenzoyl-bis(5,5-dimethylcyclohexane-1,3-dion-2-yl)methane (IIk). A solution of 22.1 g (0.1 mole) 2,4-dichlorophenylglyoxal monohydrate (Ik) and 28.0 g (0.2 mole) dimedone in a mixture of 100 ml acetic acid and 22 ml (0.35 mole) acetic anhydride was boiled for 5 min, 150 ml water was added, and the mixture was allowed to stand overnight at 20°C. The resulting white precipitate of compound IIk was removed by filtration and recrystallized from ethanol. PMR spectrum ($CDCl_3$): 0.94 (12H, s, CH_3), 2.26 (8H, s, CH_2); 5.65 (1H, s CH); 7.07-7.45 (3H, m, $C_6H_3Cl_2$); 10.70-11.80 ppm (2H OH enol band). UV spectrum (in ethanol), λ_{max} (log ε): 207 (4.46), 257 nm (4.27).

9-Aroyl-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroxanthene-1,8-diones (IIIa-n). A solution of 0.1 mole of the corresponding glyoxal Ia-n and 0.2 mole of dimedone in a mixture of 100 ml acetic acid and 47 ml (0.5 mole) acetic anhydride was boiled for 30 min, mixed with 150 ml water, and maintained for 1 h at 20°C. The resulting colorless crystalline precipitate was removed by filtration and recrystallized from ethanol or dioxane. Yields and physical characteristics of compounds IIIa-n are reported in Tables 1 and 2.

2-Phenyl-3-(6-amino-4,4-dimethyl-6-cyclohexen-2-on-1-yl)-6,6-dimethyl-4,5,6,7-tetrahydroindol-4-one (Va). A 0.1-liter volume steel autoclave at 100°C was used to heat over a 6-h period a solution of 9.8 g (0.025 mole) 9-phenyl-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroxanthene-1,8-dione (IIIa) in a mixture of 50 ml ethanol and 30 ml (0.44 mole) 25% aqueous ammonia. After being cooled the reaction mixture was treated with 30 ml water. A light yellow percipitate was deposited, which was recrystallized from ethanol and dried under vacuum for 30 min at 130°C. IR spectrum (nujol): 3400 (NH_2), 3340 (NH), 1670, 1635, and 1610 cm^{-1} (C=O). UV spectrum (in ethanol), λ_{max} (log ε): 203 (4.24), 277 (4.44), sh 350 nm (3.49). PMR spectrum (DMSO-D_6): 0.96 and 1.02 [2 × 3H, two s, 6-$C(CH_3)_2$] 1.10 [6H, s, 3-(4'-$C(CH_3)_2$]; 2.03 [4H, s, 3-(3',5'-CH_2)]; 2.57 (2H, s, 5-CH_2); 2.74 (2H, s, 7-CH_2); 7.00-7.55 (5H, m, C_6H_5); 9.75-10.70 (2H, NH_2 band); 11.85 ppm (1H, br s, 1-NH).

2-(p-Tolyl)-3-(6-amino-4,4-dimethyl-6-cyclohexen-2-on-1-yl)-6,6-dimethyl-4,5,6,7-tetrahydroindol-4-one (Vd). This was prepared from 9-(p-toluoyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroxanthene-1,8-dione (IIId) in a manner analogous to that described above for compound Va and was recrystallized from ethanol. The light yellow crystalline substance was dried under vacuum at 130°C. IR spectrum (nujol): 3630 (NH_2), 3345 (NH), 1665 cm^{-1} (C=O). UV spectrum (in ethanol), λ_{max} (log ε): 206 (4.22), 278 (4.46), sh 354 nm (3.46). PMR spectrum (DMSO-D_6): 0.97 and 1.02 [2 × 3 H, two s, 6-$C(CH_3)_2$], 1.08 [6H, s, 3-(4'-$C(CH_3)_2$]; 2.03 [2H, c, 3-(3'5'-CH_2)]; 2.25 [3H, s, 2-(4'-CH_3)]; 2.57 (2H, s, 5-CH_2); 2.72 (2H, s, 7-CH_2); 7.02 [2H, d, J = 9 Hz, 2', 6'-H)]; 7.39 [2H, d, J = 9 Hz, 2-(3',5'-H)]; 9.50-10.50 (2H, NH_2 band), 11.75 ppm (1H, br. s, 1-NH).

2-(4-Nitrophenyl)-3-(4,4-dimethyl-6-hydroxy-6-cyclohexen-2-on-1-yl)-6,6-dimethyl-4,5,6,7-tetrahydroindol-4-one (V*l*) was prepared from 9-(4-nitrobenzoyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroxanthene-1,8-dione (III*l*), in analogy with compound Va. After acifification of the reaction mixture with hydrochloric acid V*l* was isolated as a bright yellow substance which was recrystallized from ethanol and dried under vacuum at 130°C for 30 min. IR spectrum (nujol): 3520 (OH), 3460 (NH), 1630 and 1610 cm^{-1} (C=O). UV spectrum (in ethanol), λ_{max} (log ε): 202 (4.32), 244 (4.39), 388 nm (4.22). PMR spectrum (DMSO-D_6): 1.07 [6H, s, 3-(4'-$C(CH_3)_2$]; 1.09 and 1.19 [2 × 3H, two s, 6-$C(CH_3)_2$]; 2.12 (2H, s, 7-CH_2); 2.26 [4H, s, 3-(3',5'-CH_2)]; 2.69 (2H, s, 5-CH_3); 7.61 [2H, d, J = 9.0 Hz, 2-(2',6'-H)]; 8.16 [2H, d, J = 9.0 Hz, 2-(3',5'-H)]; 10.02 (1H, br.s, 2-NH); 11.77 ppm (1H, s, OH).

1-Aryl-4,4,8,8-tetramethyl-2,3,4,5,7,8,9,10-octahydropyrrolo[4,3,2-m,n]acridin-10-ones (VIa-m). A. A solution of the appropriate aroylbis(5,5-dimethylcyclohexan-1,3-dion-2-yl)methane derivative (IIa-m) (0.025 mole) or 9-aroyl-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroxanthene-1,8-dione (IIIa-m) in a mixture of 50 ml ethanol and 30 ml (0.44 mole) 25% aqueous ammonia was heated in a steel autoclave at 130-150°C for 5 h. After being cooled the reaction mixture deposited yellow or orange crystalline substances VIa-m, which were recrystallized from ethanol or a mixture of ethanol and water (7:3) and subsequently dried under vacuum at room temperature. Solvents of crystallization were removed by heating under vacuum at 80-130°C. Yields and physical characteristics of these compounds are presented in Tables 3-5.

B. A solution of 0.2 g (0.5 mmole) compound V*l* in 1 ml ethanol and 0.7 ml (10 mmole) 25% aqueous ammonia was heated at 130°C for 2 h in a sealed, thick-walled 5-ml-volume glass reaction flask (Reacti-Vial TM), allowing solution color changes to be observed. After cooling, 0.1 g (50%) of 1-(4-nitrophenyl)-4,4,8,8-tetramethyl-2,3,4,5,7,8,9,10-octahydropyrrolo[4,3,2-m,n]acridin-10-one (VI*l*) was obtained, which was similar to the material prepared as in method A above.

*Hydrochloride Salt, 1-(4-Nitrophenyl)-4,4,8,8-tetramethyl-2,3,4,5,7,8,9,10-octahydropyrrolo[4,3,2-m,n]acridin-10-one (VII*l*).* This was prepared by addition of 0.25 ml (2.5 mmole) of conc. HCl to a solution of 1.0 g (2.4 mmole) acridine VII in 25 ml hot ethanol. After cooling 0.9 g of a bright yellow material was obtained, which was recrystallized from a mixture of ethanol and water.

Hydrochloride Salt, 1-Phenyl-4,4,8,8-tetramethyl-2,3,4,5,7,8,9,10-octahydropyrrolo[4,3,2-m,n]acridin-10-one (VIIa). This was prepared by analogy with salt VII*l*. Light orange substance, which was recrystallized from a mixture of ethanol and water.

*Potassium salt, 1-(4-Nitrophenyl)-4,4,8,8-tetramethyl-2,3,4,5,7,8,9,10-octahydropyrrolo[4,3,2-m,n]acridin-10-one (VIII*l*).* A dark violet solution of 2.0 g (4.7 mmole) 1-(4-nitrophenyl)-4,4,8,8-tetramethyl-2,3,4,5,7,8,9,10-octahydropyrrolo[4,3,2-m,n]acridin-10-one (VI*l*) in 5 ml ethanol containing 0.8 g (14 mmole) potassium hydroxide deposited a dark orange crystalline precipitate. Hexane (5 ml) was added and the precipitate was filtered, washed with hexane, and dried at room temperature. Yield 1.9 g (85%). Potentiometric titration of the potassium salt VIII*l* with a solution of hydrochloric acid in alcohol (accompanied by a color change to orange) indicated a 95% concentration of potassium ion when calculated based on a dihydrate of salt VIII*l*. (These measurements were made by the analytical chemist G. E. Krastinei.)

Reaction of 9-(2-Nitrobenzoyl)-3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroxanthene-1,8-dione (IIIn) with Ammonia. A solution of 0.2 g (0.51 mmole) dione IIIn in a mixture of 2 ml ethanol and 1.0 ml (15 mmole) 25% aqueous ammonia was heated at 120°C for 30 min in a sealed, thick-walled glass reaction flask (5 ml volume). After cooling 0.1 g (47%) of a light orange substance was obtained, which was recrystallized from dioxane. The product was a light yellow crystalline material, 4-(2-nitrophenyl)3,3,6,6-tetramethyl-1,2,3,4,5,6,7,8-octahydroacridine-1,8-dione, mp 277-280°C. When the reaction was carried out under harsher conditions, resinification of the reaction mixture was observed. UV spectrum (in ethanol), λ_{max} (log ϵ): 205 (4.89), sh 243 (4.63), 293 (4.13), sh 302 (4.08), sh 340 nm (3.19). PMR spectrum ($CDCl_3$): 1.12 and 1.13 [2 × 6H, two s, 3,6-$C(CH_3)_2$], 2.50 (4H, s, 2.7-CH_2); 3.08 (4H, s, 4,5-CH_2); 7.32-7.68 ppm (4H, n, C_6H_4). Found: C 69.2; H 5.9; N 6.9%; M^+ 420. $C_{24}H_{24}N_2O_6$. Calc.: C 68.6, H 5.8; N 6.7%; M 420.

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