

# Some 2,6-di(*p*-X-phenyl)-4,4-diphenyl-4*H*-pyrans and analogous 1,4-dihydropyridines from 1,5-dione precursors

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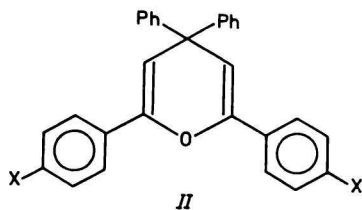
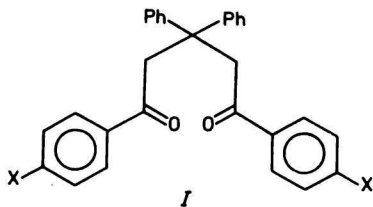
*Dedicated to Professor Ing. J. Kováč, DrSc., in honour of his 60th birthday*

Substituted 1,3,3,5-tetraphenyl-1,5-pentadiones *Ia—Id*, obtained by alkaline ketolization of the corresponding acetophenones with benzophenone, have been subjected to intramolecular cyclocondensation to give 4*H*-pyrans *IIa—IId* and 1,4-dihydropyridines *IIIa—IIIf*. Mass spectrometric fragmentation of the above-mentioned compounds is discussed. The compounds *Ia, Ib, IIb—IId, IIIa—IIIf* in crystalline state showed photochromy.

Замещенные 1,3,3,5-тетрафенил-1,5-пентадионы *Ia—Id*, полученные в результате щелочной кетолизации соответствующих ацетофенонов с бензофеноном, были превращены посредством внутримолекулярной циклоконденсации в 4*H*-пираны *IIa—IId* и 1,4-дигидропиридины *IIIa—IIIf*. Обсуждается масс-спектрометрическая фрагментация данных соединений. Соединения *Ia, Ib, IIb—IId, IIIa—IIIf* проявляют фотохромиию в кристаллическом состоянии.

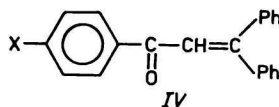
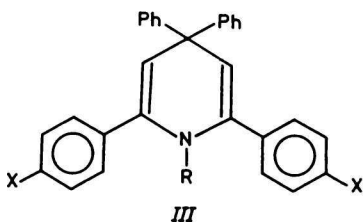
Some time ago *Carvalho* [1—3] described ketolization of acetophenone with benzophenone by treatment with sodium amide under formation of 1,3,3,5-tetraphenyl-1,5-pentadione (*Ia*; see Formula 1) which by treatment with phosphoric oxide and ammonium acetate, respectively, cyclized to 2,4,4,6-tetraphenyl-4*H*-pyran (*IIa*) and 2,4,4,6-tetraphenyl-1,4-dihydropyridine (*IIIa*), respectively. The author [2, 3] observed significant photochromy when he exposed the compounds *Ia* and *IIIa* to sunlight. Studies of photochromic properties of *IIIa* in solutions revealed [4] that in the presence of oxygen photochromy did not occur. Also sensitized photooxidation of *IIIa* was studied [5]. In connection with our interest in compounds having defined optical properties we were interested in how photochromy and stability of the compounds investigated would be influenced by introducing a *para*-substituent into terminal phenyl groups of precursors of the type *I* and also by a possible substitution on nitrogen

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X

- a H
- b CH<sub>3</sub>
- c OCH<sub>3</sub>
- d Br



X

- a H
- b OCH<sub>3</sub>

	R	X
a	H	H
b	CH <sub>3</sub>	H
c	Ph	H
d	CH <sub>2</sub> Ph	H
e	CH <sub>3</sub>	CH <sub>3</sub>
f	CH <sub>3</sub>	Br

atom in heterocycles of the type *III*. Therefore, we reproduced the cited [1—3, 6] preparation of compounds *Ia*, *Ila*, *IIIa* and, moreover, prepared analogous compounds *Ib—Id*, *Ilb—Ild*, and *IIIb—III f*. Besides, we investigated the behaviour of molecules of the prepared compounds *II* and *III* in gaseous state under electron impact at 70 eV. The results obtained are the subject of the present work.

From the results obtained in condensation of benzophenone with acetophenone by using sodium ethoxide, sodium hydride, and sodium amide it follows that satisfactory yield of diketone *Ia* was achieved only by action of sodium amide. In the reaction mixture a further product, 1,3,3-triphenyl-2-propanone (*Iva*), was identified. This is evidently an intermediate of the reaction. Obviously, sodium amide spread up both reaction steps most effectively.

This experience was utilized in analogous preparation of substituted 1,5-pentadiones *Ib—Id* by the reaction of benzophenone with *p*-substituted acetophenones in the presence of sodium amide. From the data in Table 1 it is evident that the yields of 1,5-diketones *I* increase with the increasing values of  $\sigma$ -constant [7] of the substituent *X*, in accordance with increase of prototropic activity of the methyl group in *p*-substituted acetophenones. The highest yields of 2,4,4,6-tetraphenyl-4*H*-pyran (*Iia*) were achieved in the reaction of diketone *Ia* with phosphoric oxide, while acetic anhydride with trace amount of acetyl chloride, iodine or *p*-toluenesulfonic acid gave lower yields of *Iia*. The substituted 4*H*-pyrans *Iib—Iid* were prepared analogously by treatment of diketones *Ib—Id* with phosphoric oxide.

2,4,4,6-Tetraphenyl-1,4-dihydropyridine (*IIIa*) was obtained only by treatment of diketone *Ia* in glacial acetic acid with ammonium acetate. When introducing ammonia into the solution of diketone *Ia* or on heating *Ia* in formamide only the starting compound *Ia* was recovered from the reaction mixture. In order to investigate the stability of the 2,4,4,6-tetraphenyl-1,4-dihydropyridine skeleton closer, *N*-substituted 1,4-dihydropyridines *IIIb—IIIid* were prepared by cyclocondensation of diketone *Ia* with acetates of the respective primary amines in acetic acid. The yields of the compounds *IIIb—IIIid* (Table 1) decrease in the order of methyl, benzyl, and phenyl derivatives, *i.e.* with the increasing sterical requirements of the substituents. It has been observed that *N*-substitution has favourable effect on stability of the obtained heterocycles. Further, we attempted to prepare 1,4-dihydropyridine derivatives *III* substituted in *para*-positions of 2,6-phenyl groups from the corresponding 1,5-diketones *Ib—Id* by the reaction with methylammonium acetate. Contrary to bis-methylphenyl and bis-bromophenyl derivatives *Ib* and *Id*, where heterocyclic ring closure proceeds with satisfactory yields, bis-methoxyphenyl derivative *Ic* does not undergo the above-mentioned cyclization. In this case besides the unreacted starting compound *Ic* only 3,3-diphenyl-1-(*p*-methoxyphenyl)-2-propenone (*IVb*) was isolated. Its formation can probably be explained by retroketolization to *p*-methoxyacetophenone and unsaturated ketone. This splitting of the diketone molecule may be a consequence of slower nucleophilic attack of its carbonyl groups due to the effect of the *para*-methoxy substituent.

The prepared compounds were characterized by elemental analysis and melting points (Table 1). Their structures were proved by IR and <sup>1</sup>H NMR spectra (Tables 2 and 3). The IR spectra of 1,5-diketones *I* showed an intense absorption band in the region of  $\tilde{\nu} = 1680—1685 \text{ cm}^{-1}$ , characteristic of aromatic ketones [8]. The absorption bands at  $\tilde{\nu} = 1630—1690 \text{ cm}^{-1}$  and  $\tilde{\nu} = 1180—1220 \text{ cm}^{-1}$  [9], belonging to stretching vibrations of the C=C and C—O bonds, are characteristic of 4*H*-pyrans *II*. The skeleton vibrations in the region of  $\tilde{\nu} = 1570—1680 \text{ cm}^{-1}$  are characteristic of 1,4-dihydropyridines *III* [10, 11].

Table 1

Characterization of the prepared compounds I—III

Compound	Formula	$M_r$	$w_i(\text{calc.})/\%$				Procedure <sup>a</sup>	Yield %	M.p. °C	Photochromy <sup>b</sup>
			$w_i(\text{found})/\%$							
			C	H	Br	N				
<i>Ia</i>	$\text{C}_{29}\text{H}_{24}\text{O}_2$	404.5134	86.14	5.94			<i>A</i>	2	187—188	Violet
			86.23	6.06			<i>B</i>	3		
							<i>C</i>	39		
							<i>D</i>	20		
<i>Ib</i>	$\text{C}_{31}\text{H}_{28}\text{O}_2$	432.5676	86.06	6.53			<i>C</i>	21	114—116	Red
			86.07	6.64						
<i>Ic</i>	$\text{C}_{31}\text{H}_{28}\text{O}_4$	464.5664	80.15	6.07			<i>C</i>	18	156—157	
			80.08	5.98						
<i>Id</i>	$\text{C}_{29}\text{H}_{22}\text{O}_2\text{Br}_2$	562.3155	61.94	3.92	28.44		<i>C</i>	45	195—196	
			62.02	4.05	28.61					
<i>IIa</i>	$\text{C}_{29}\text{H}_{22}\text{O}$	386.4981	90.12	5.74			<i>A</i>	57	172—173	
			90.33	5.85			<i>B</i>	44		
							<i>C</i>	47		
<i>IIb</i>	$\text{C}_{31}\text{H}_{26}\text{O}$	415.9844	89.92	6.32			<i>D</i>	52	188—189	Blue
			89.70	6.36			<i>A</i>	54		
<i>IIc</i>	$\text{C}_{31}\text{H}_{26}\text{O}_3$	446.5512	83.38	5.87			<i>A</i>	41	215—217	Blue
			83.30	5.89						

Table 1 (Continued)

Compound	Formula	$M_r$	$w_i(\text{calc.})/\%$ $w_i(\text{found})/\%$				Procedure <sup>a</sup>	Yield %	M.p. °C	Photochromy <sup>b</sup>
			C	H	Br	N				
<i>IId</i>	$C_{29}H_{20}OBr_2$	544.3002	63.99	3.70	29.36		<i>A</i>	58	262—263	Blue
			64.19	3.70	29.18					
<i>IIIa</i>	$C_{29}H_{23}N$	385.5134				<i>C</i>	83	233—235	Violet	
<i>IIIb</i>	$C_{30}H_{25}N$	399.5405	90.19	6.31		3.50	<i>C</i>	74	182—184	Violet
			90.15	6.31		3.57				
<i>IIIc</i>	$C_{35}H_{27}N$	461.6121	91.07	5.90		3.07	<i>C</i>	33	168—170	Violet
			90.98	5.83		3.14				
<i>IIId</i>	$C_{36}H_{29}N$	475.6392	90.91	6.14		2.95	<i>C</i>	66	176—178	Violet
			90.73	6.23		3.03				
<i>IIIe</i>	$C_{32}H_{29}N$	427.5946	89.89	6.84		3.27	<i>C</i>	37	212—214	Bluish-green
			89.90	6.93		3.14				
<i>IIIf</i>	$C_{30}H_{23}NBr_2$	557.3425	64.66	4.16	28.67	2.51	<i>C</i>	66	218—221	Violet
			64.72	4.12	28.78	2.38				

a) See Experimental; b) the colour of crystals after irradiation by sunlight.

Table 2

Significant absorption bands in infrared spectra of the compounds *I*–*III*

Compound	$\tilde{\nu}/\text{cm}^{-1}$		
	C = O	C = C	C–H <sub>ar</sub>
<i>Ia</i> <sup>a</sup>	1685	1594	3063
<i>Ib</i> <sup>a</sup>	1683	1607	3063
<i>Ic</i> <sup>a</sup>	1680	1600	3062
<i>Id</i> <sup>a</sup>	1685	1589	3083

Compound	$\tilde{\nu}/\text{cm}^{-1}$				
	Skeleton vibration			C–H <sub>ar</sub>	
<i>IIa</i> <sup>b</sup>	1687	1640	1212	1180	3078
<i>IIb</i> <sup>b</sup>	1679	1638	1210	1181	3067
<i>IIc</i> <sup>b</sup>	1684	1640	1205	1183	3060
<i>IIa</i> <sup>b</sup>	1685	1640	1223	1182	3060
<i>IIIa</i> <sup>a</sup>		1672	1600	1578	3038
<i>IIIb</i> <sup>b</sup>		1663	1602	1579	3072
<i>IIIc</i> <sup>b</sup>		1682	1600	1578	3070
<i>IIIa</i> <sup>b</sup>		1660	1600	1577	3079
<i>IIIe</i> <sup>b</sup>		1660	1598	1576	3060
<i>IIIf</i> <sup>b</sup>		1661	1610	1588	3060

a) Measured in KBr pellet; b) measured in CCl<sub>4</sub> solution.

The <sup>1</sup>H NMR spectra of the compounds *I*–*III* (Table 3) revealed besides the signals of aromatic hydrogen atoms and signals of the substituents the characteristic singlets of methylene groups (diketones *I*) and singlets of methine groups [10, 12] in positions 3 and 5 of 4*H*-pyran (compounds *II*) or 1,4-dihydropyridine (compounds *III*) ring. Integral intensities and the numbers of signals show that the compounds *I* occur practically only in the diketo form under the conditions of measurements (trifluoroacetic acid).

Analysis of the mass spectra of 4*H*-pyrans *II* and 1,4-dihydropyridines *III* showed that under electron impact both groups of compounds underwent similar conversions. Typical fragmentation mechanism of 4*H*-pyrans *II* is illustrated in Scheme 1 and of 1,4-dihydropyridines *III* in Scheme 2. The basic splitting is the typical [10, 11] aromatization of the molecular ion under forma-

Table 3

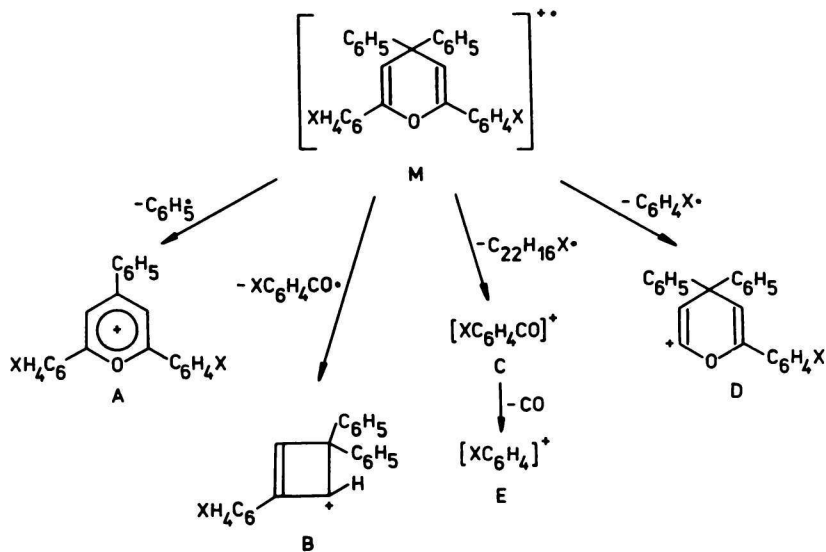
<sup>1</sup>H NMR spectra of the compounds I—III

Compound	$\delta$ /ppm			
	(CH <sub>2</sub> )	(CH) <sup>a</sup>	(CH) <sup>b</sup>	Others
<i>Ia</i> <sup>c</sup>	4.33 (s)	7.04—7.83 (m)		
<i>Ib</i> <sup>c</sup>	4.21 (s)	7.10—7.37 (m)	7.20 (d), 7.72 (d) <sup>3</sup> J <sub>HH</sub> = 8 Hz	2.43 (s, CH <sub>3</sub> )
<i>Ic</i> <sup>c</sup>	4.10 (s)	7.10—7.34 (m)	6.85 (d), 7.84 (d) <sup>3</sup> J <sub>HH</sub> = 9 Hz	3.95 (s, CH <sub>3</sub> )
<i>Id</i> <sup>c</sup>	4.32 (s)	7.05—7.37 (m)	7.52 (s)	
Compound	(=CH—)	(CH) <sup>a</sup>	(CH) <sup>b</sup>	Others
<i>IIa</i> <sup>d</sup>	5.71 (s)	7.00—7.64 (m)		
<i>IIb</i> <sup>d</sup>	5.79 (s)	7.05—7.46 (m)	6.97 (d), 7.63 (d) <sup>3</sup> J <sub>HH</sub> = 8 Hz	2.12 (s, CH <sub>3</sub> )
<i>IIc</i> <sup>d</sup>	5.70 (s)	6.88—7.47 (m)	6.74 (d), 7.60 (d) <sup>3</sup> J <sub>HH</sub> = 9 Hz	3.30 (s, OCH <sub>3</sub> )
<i>IIId</i> <sup>d</sup>	5.59 (s)	6.92—7.35 (m) <sup>e</sup>		
<i>IIIb</i> <sup>f</sup>	5.20 (s)	6.96—7.60 (m) <sup>e</sup>		2.58 (s, NCH <sub>3</sub> )
<i>IIIc</i> <sup>f</sup>	5.65 (s)	7.07—7.77 (m) <sup>e</sup>		
<i>IIId</i> <sup>f</sup>	5.16 (s)	6.41—7.58 (m) <sup>e</sup>		4.20 (s, CH <sub>2</sub> )
<i>IIIe</i> <sup>f</sup>	5.12 (s)	7.10—7.32 (m)	7.06 (d), 7.38 (d) <sup>3</sup> J <sub>HH</sub> = 8 Hz	2.35 (s, CH <sub>3</sub> )
<i>IIIff</i>	5.10 (s)	7.10—7.56 (m) <sup>e</sup>		2.57 (s, NCH <sub>3</sub> ) 2.28 (s, NCH <sub>3</sub> )

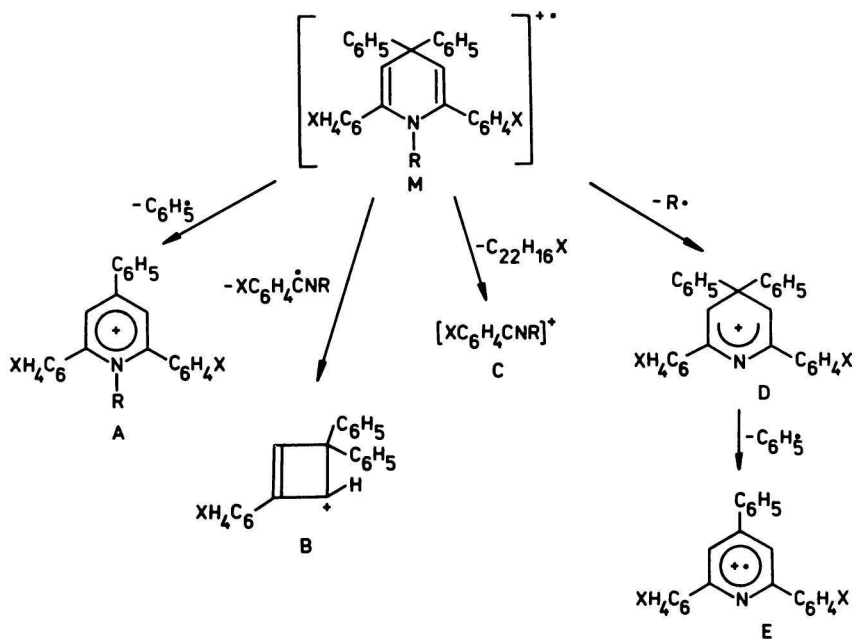
*a*) Unsubstituted phenyls; *b*) substituted phenyls; *c*) measured in CF<sub>3</sub>COOH; *d*) measured in C<sub>6</sub>D<sub>6</sub>; *e*) the signals are undistinguishable; *f*) measured in CCl<sub>4</sub>.

tion of pyrylium and pyridinium ions A (Table 4) where phenyl is split off from the position 4. The ions B and C (Schemes 1 and 2) are formed by fission of the heterocyclic ring. The ions D and E and their formation are characteristic [10, 12] only of the individual groups of compounds *II* and *III*. The suggested fragmentation mechanisms (Schemes 1 and 2; Table 4) were confirmed by the presence of the corresponding metastable ions.

In addition to compounds *Ia* and *IIIa*, the photochromic properties of which were already mentioned, majority of the newly prepared compounds *I*, *II*, and *III* exhibited photochromic behaviour in the visible region. When exposed to



Scheme 1



Scheme 2



Table 4

Chosen characteristic ions in mass spectra of the compounds *II*  
(Scheme 1) and *III* (Scheme 2)

Compound	$m/z$ ( $I_r$ ) Ion <sup>a</sup>					
	M	A	B	C	D	E
<i>IIa</i>	386(50)	309(100)	281(26)	105(57)	309(100)	77(36)
<i>IIb</i>	414(55)	337(100)	295(35)	119(80)	323( 5)	91(45)
<i>IIc</i>	446(22)	369(100)	311( 8)	135(39)	339( 11)	107( 8)
<i>IId</i>	542(27)	465( 54)	359(27)	183(54)	387( 16)	155(27)
	544(54)	467(100)	361(24)	185(54)	389( 16)	157(27)
	546(27)	469( 24)				
<i>IIIa</i>	385(50)	308(100)	281( 6)	104(65)	384( 11)	307(19)
<i>IIIb</i>	399(22)	322(100)	281( 2)	118( 3)	384( 2)	307(21)
<i>IIIc</i>	461(34)	384(100)	281(80)	180( 2)	384(100)	307( 3)
<i>IIId</i>	475(67)	398(100)	281( 8)	194( 4)	384( 31)	307(11)
<i>IIIe</i>	427(24)	350(100)	295( 3)	132( 2)	412( 2)	335( 3)
<i>IIIf</i>	553(25)	476( 82)	359( 2)	196( 7)	540( 2)	463( 2)
	555(50)	478(100)				
	557(25)	480( 82)				

a) The structures of the individual ions are illustrated in Schemes 1 and 2.

UV irradiation the colourless crystals of these compounds changed colour (Table 1). On irradiation of the sample by sunlight the time necessary for exposure varied from seconds to several minutes. On allowing the colour crystals to stay in the dark, the colour was observed to be lost within several hours to weeks. Photochromic phenomena observed with compounds *I*—*III* cannot be explained by the known [13] mechanism of photochromism of organic compounds. These problems are subject to further investigation.

## Experimental

Melting points were determined on a Kofler block. IR spectra were recorded with a Perkin—Elmer 325 spectrophotometer, mass spectra were measured with an LKB 9000 (70 eV, direct inlet) spectrometer, and <sup>1</sup>H NMR spectra with a Varian 100 XL apparatus (internal standard TMS).

## 1,5-Diketones I

### Procedure A

Sodium ethoxide (160 mmol) was layered with the solution of acetophenone (75 mmol) and benzophenone (37 mmol) in benzene (50 cm<sup>3</sup>) and boiled for 8 h using an azeotropic water separator. After decomposition with water, the starting compounds were distilled off from the reaction mixture. From the distillation residue the product *Ia* was precipitated with diethyl ether.

### Procedure B

The mixture of sodium hydride (210 mmol), benzophenone (50 mmol), and acetophenone or *p*-substituted acetophenone (100 mmol) in ether (50 cm<sup>3</sup>) was stirred for 4 days at room temperature. After decomposition with ethanol and water, the starting compounds were distilled off with water vapours. The distillation residue was extracted with benzene which was distilled off after drying with anhydrous magnesium sulfate. The product *Ia* was obtained on layering the residue with ether.

### Procedure C

The mixture of sodium amide (200 mmol), benzophenone (50 mmol), and *p*-substituted acetophenone (100 mmol) in ether (50 cm<sup>3</sup>) was stirred for 3 h at 0 °C and then for 2 days at room temperature. After decomposition with water and extraction of aqueous phase with benzene, the organic portions were dried with anhydrous magnesium sulfate. The crude product, obtained on removal of solvents by distillation, was distilled with water vapours and the distillation residue was extracted with benzene. After drying, evaporation of benzene, and layering with ether the products *Ia*—*Id* were obtained.

### Procedure D

The mixture of sodium amide (21 mmol), 1,3,3-triphenyl-2-propenone (7 mmol), and acetophenone (7 mmol) in ether (20 cm<sup>3</sup>) was stirred for 4 days at room temperature. The reaction mixture was decomposed with water, neutralized with diluted hydrochloric acid, and extracted with benzene. After drying of organic portions and removal of solvents by distillation, the product *Ia* was precipitated with hot methanol.

## 4H-Pyrans II

### Procedure A

All operations were performed in the atmosphere of nitrogen, dried over phosphoric oxide. The mixture of pentadione *Ia* and phosphoric oxide (27.2 mmol) was layered with

xylene (20 cm<sup>3</sup>) and boiled for 3 h. After cooling the xylene solution was decanted and evaporated *in vacuo*. From the distillation residue the crude product *IIa* was precipitated with petroleum ether and crystallized from the mixture of benzene and petroleum ether.

#### *Procedure B*

The solution of diketone *Ia* (2.7 mmol), acetyl chloride (0.1 cm<sup>3</sup>), and acetic anhydride (10 cm<sup>3</sup>) was boiled for 10 h. After cooling the obtained crystals of the crude product *IIa* were purified similarly as in the procedure *A*.

#### *Procedure C*

From the solution of diketone *Ia* (2.6 mmol) and iodine (0.8 mmol) in toluene (180 cm<sup>3</sup>) the azeotropic mixture of toluene and water was distilled off during 30 h. The crude product *IIa*, obtained from the distillation residue, was purified as in the preceding procedures.

#### *Procedure D*

From the mixture of diketone *Ia* (2.7 mmol), *p*-toluenesulfonic acid (0.3 mmol), and toluene (180 cm<sup>3</sup>) the azeotropic mixture of toluene and water was distilled off within 30 h. The crude product *IIa*, obtained from the distillation residue, was purified as in the preceding procedures.

### *1,4-Dihydropyridines III*

#### *Procedure A*

Into the solution of pentadione *Ia* (10 mmol) in benzene (150 cm<sup>3</sup>) ammonia dried with sodium hydroxide was introduced at 20°C during 15 h. After evaporation of benzene, only the starting diketone *Ia* recovered.

#### *Procedure B*

The solution of diketone *Ia* (5 mmol) in formamide (15 cm<sup>3</sup>) was boiled for 1 h. After cooling and dilution with water, only the starting diketone *Ia* was recovered.

#### *Procedure C*

The mixture of diketone *I* (5 mmol) and 4 to 15-fold excess of the corresponding ammonium acetate in glacial acetic acid (30 to 100 cm<sup>3</sup>) was boiled for 2 to 20 h. The

crude crystalline product obtained after cooling was recrystallized from acetone. When the product after cooling did not crystallize, then after dilution with water, extraction with benzene, drying the benzene phase with calcium chloride, and evaporation of benzene, the mixture of the starting compound *I* and the product *III* was obtained. This mixture was separated by column chromatography on silica gel (eluent tetrachloromethane or the solution of 10 % chloroform in tetrachloromethane). In the case of diketone *Ic* elution with chloroform gave 0.40 g (25 % yield) of substituted chalcone *IVb*. Recrystallization from methanol gave the product of m.p. = 101–102 °C. Ref. [14] gives m.p. = 105 °C.

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