

## SYNTHESIS AND THE PROPERTIES OF FURFURYLIDENE- AND THENYLIDENEINDOXYLS\*

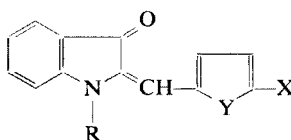
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2-(5-X-2-furfurylidene)indoxyls ( $X = \text{H}, \text{NO}_2, \text{Cl}, \text{Br}, \text{I}, \text{CH}_3, \text{SCH}_3, \text{COOH}$ ) and 2-(5-X-2-thenylidene)indoxyls ( $X = \text{H}, \text{NO}_2$ ) were prepared by hydrolysis of corresponding 1-acetyl derivatives, condensation products of 1-acetylindoxyl with 5-X-2-furan- and 2-thiophenecarbaldehydes, or by reacting 3-acetylindoxyl with the mentioned aldehydes. The infrared, ultraviolet and mass spectra of the final products were measured.

In view of the knowledge that some furfurylideneoxindoles with an exocyclic double bond on the  $C_{(3)}$  atom of oxindol, among them in the first place 3-(5-nitro-2-furfurylidene)oxindole, possess considerable antibacterial and antiseptic properties<sup>1-7</sup>, the question of the synthesis and the study of the properties of analogous derivatives of indoxyl as yet undescribed in literature, such as furfurylideneindoxyls or thenylideneindoxyls, seemed interesting. In this paper a simple synthesis and physico-chemical properties of substituted 2-(2-furfurylidene- or 2-thenylidene)indoxyls I–XVIII is described.



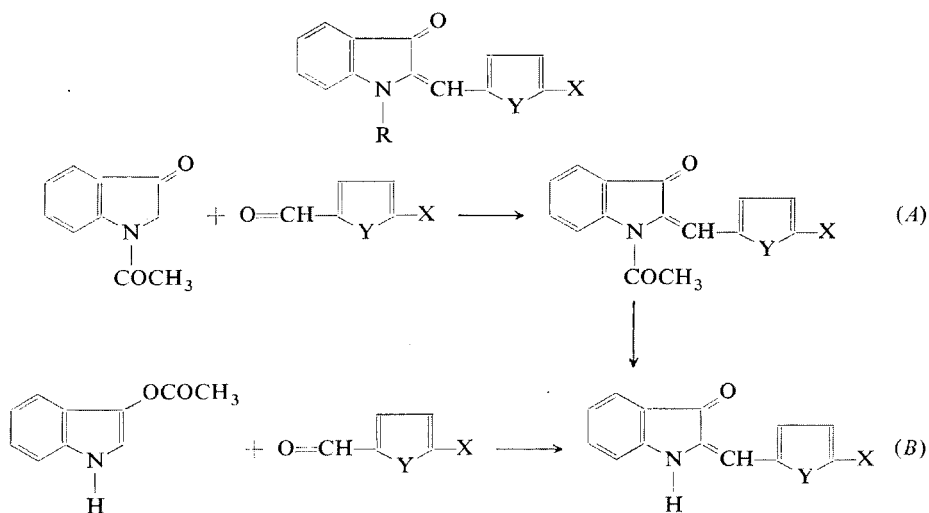
	R	Y	X		R	Y	X
I	COCH <sub>3</sub>	O	H	X	H	O	NO <sub>2</sub>
II	COCH <sub>3</sub>	O	NO <sub>2</sub>	XI	H	O	Cl
III	COCH <sub>3</sub>	O	CH <sub>3</sub>	XII	H	O	Br
IV	COCH <sub>3</sub>	O	SCH <sub>3</sub>	XIII	H	O	I
V	COCH <sub>3</sub>	O	COOCH <sub>3</sub>	XIV	H	O	CH <sub>3</sub>
VI	COCH <sub>3</sub>	O	N(CH <sub>3</sub> ) <sub>2</sub>	XV	H	O	SCH <sub>3</sub>
VII	COCH <sub>3</sub>	S	H	XVI	H	O	COOH
VIII	COCH <sub>3</sub>	S	NO <sub>2</sub>	XVII	H	S	H
IX	H	O	H	XVIII	H	S	NO <sub>2</sub>

\* Part LIX in the series Furan Derivatives; Part LVIII: This Journal 40, 1563 (1975).

The classical method of formation of arylidene- and alkylideneindoxyls, the so-called "indogenins", based on the reaction of indoxyl<sup>8,9</sup>, indoxyl acid<sup>8</sup>, or indole oxidized with air oxygen<sup>10</sup>, with carbonyl compounds did not give good results in the preparation of 2-(5-X-2-furfurylidene- and 2-thenylidene)indoxyls *IX–XVIII*. This circumstance is also connected with the low stability of indoxyl, which undergoes oxidative dimerization to indigo exceptionally easily. For the preparation of the compounds of this type more stable derivatives of indoxyl, such as 1-acetylindoxyl (Method *A*) and 3-acetylindoxyl (Method *B*) (Scheme 1), were used successfully. The carbonyl components of the mentioned condensation reactions were 5-X-2-furancarbaldehydes ( $X = \text{H}, \text{NO}_2, \text{Cl}, \text{Br}, \text{I}, \text{CH}_3, \text{SCH}_3, \text{COOCH}_3, \text{N}(\text{CH}_3)_2$  and 5-X-2-thiophenecarbaldehydes ( $X = \text{H}, \text{NO}_2$ ).

From anthranilic and chloroacetic acids potassium salt of *o*-carboxyphenylglycine<sup>11</sup> was prepared which was then converted to 1-acetylindoxyl<sup>13</sup> by cyclization to 1,3-diacetylindoxyl<sup>12</sup> and subsequent partial hydrolysis. 3-Acetylindoxyl was obtained by a two-step synthesis from indole (via 3-iodoindole<sup>14</sup>).

On condensation of equimolar amounts of 5-X-2-furancarbaldehydes ( $X = \text{H}, \text{NO}_2, \text{CH}_3, \text{SCH}_3, \text{COOCH}_3, \text{N}(\text{CH}_3)_2$ ) and 5-X-2-thiophenecarbaldehydes ( $X = \text{H}, \text{NO}_2$ ) with 1-acetylindoxyl in benzene under catalysis with piperidine 1-acetyl-2-(5-X-2-furfurylidene- or 2-thenylidene)indoxyls *I–VIII* were prepared (Method *A*; see Table I). The formation of 2-(5-X-2-furfurylidene- or 2-thenylidene)indoxyls by acid hydrolysis of acetyl derivatives *I–VIII* was successful only in the case of synthesis of compounds *IX, X, XVII* and *XVIII*. The hydrolysis was carried out by gradual addition of dilute hydrochloric acid into boiling 1-acetyl derivative dissolved



SCHEME 1

in a water-miscible solvent (methanol, ethanol, propanol, acetone). The yields of these reactions were 47–72% (Table II).

Applying the method of Abramovitch and Marko<sup>15</sup> much higher yields were achieved (52–95%) in the synthesis of 2-(5-X-2-furfurylidene)indoxyl IX, XI–XVI. The method is based on the action of 3-acetylindoxyl on benzenecarbaldehydes in aqueous solution of alkali hydroxides and subsequent extraction of the reaction mixture with a suitable solvent, mostly ether (Method B, see Table II).

Both methods of synthesis of compounds IX–XVIII were suitably combined. Compounds XIII, XV and XVI could not be prepared by acid hydrolysis of corresponding 1-acetyl derivatives; however, the mentioned compounds are formed in good yield from 3-acetylindoxyl. In contrast to this alkaline medium has such an unfavourable effect on 5-nitro-2-furancarbaldehyde that the synthesis of X can be achieved by Method A only. Compound IX, prepared by both methods, was identical.

TABLE I

1-Acetyl-2-(5-X-2-Furfurylidene-, or 2-Thenylidene)indoxyls I–VIII  
(R = COCH<sub>3</sub>)

Compound	Composition (m.w.)	Calculated/found			M.p., °C (yield, %)	Solvent
		% C	% H	% N		
I	C <sub>15</sub> H <sub>11</sub> NO <sub>3</sub> (253.3)	71.13	4.38	5.53	90–93	cyclohexane
		71.28	4.35	5.51	(51.4)	
II	C <sub>15</sub> H <sub>10</sub> N <sub>2</sub> O <sub>5</sub> (298.3)	60.40	3.38	9.39	191–192.5	benzene
		60.20	3.24	9.46	(76.8)	
III	C <sub>16</sub> H <sub>13</sub> NO <sub>3</sub> (267.3)	71.89	4.90	5.24	122–124.5	70% ethanol
		71.82	4.93	5.25	(45.0)	
IV	C <sub>16</sub> H <sub>13</sub> NO <sub>3</sub> S <sup>a</sup> (299.3)	64.19	4.38	4.68	105–107	70% ethanol
		64.22	4.40	4.59	(30.1)	
V	C <sub>17</sub> H <sub>13</sub> NO <sub>5</sub> (311.3)	65.59	4.21	4.50	170–173	ethanol
		65.21	4.18	4.22	(28.9)	
VI	C <sub>17</sub> H <sub>16</sub> N <sub>2</sub> O <sub>3</sub> (296.3)	68.90	5.44	9.46	133–135	benzene–cyclohexane (2 : 1)
		68.75	5.40	9.34	(88.1)	
VII	C <sub>15</sub> H <sub>11</sub> NO <sub>2</sub> S <sup>b</sup> (269.3)	66.89	4.11	5.20	130–131	ethanol
		66.84	4.08	5.28	(74.3)	
VIII	C <sub>15</sub> H <sub>10</sub> N <sub>2</sub> O <sub>4</sub> S <sup>c</sup> (314.3)	57.31	3.21	8.91	217–219	acetone
		57.05	3.18	8.90	(38.2)	

<sup>a</sup> Calculated 10.71% S, found 10.75% S; <sup>b</sup> calculated 11.91% S, found 11.87% S; <sup>c</sup> calculated 10.20% S, found 10.12% S.

The synthesis of 2-(5-dimethylamino-2-furfurylidene)indoxyl could not be carried out by either of these methods. Both methods of synthesis of 2-(5-X-2-furfurylidene or 2-thenylidene)indoxyls eliminate the difficulties connected with the oxidation of indoxyl to indigo. Method *A* does not give this by-product at all. A part of indigo formed by method *B* can be easily separated from the final product.

In the IR absorption spectra of compounds *I–XVIII* (Table III) C=O stretching vibrations in the 1722–1665 cm<sup>-1</sup> region can be observed. In the case of 1-acetyl derivatives *I–VIII* the band with higher frequency (1716–1689 cm<sup>-1</sup>) corresponds

TABLE II  
2-(5-X-2-Furfurylidene, or 2-Thenylidene)indoxyls *IX–XVIII* (R = H)

Compound	Composition (m.w.)	Calculated/found			M.p., °C (yield, %)	Solvent
		% C	% H	% N		
<i>IX</i>	C <sub>13</sub> H <sub>9</sub> NO <sub>2</sub> (211·21)	73·92	4·29	6·63	171–173 (66·6)	cyclohexane
		73·87	4·19	6·66		
<i>X</i>	C <sub>13</sub> H <sub>8</sub> N <sub>2</sub> O <sub>4</sub> (256·21)	60·94	3·15	10·94	232–234 (47·0)	ethanol
		61·12	3·15	10·95		
<i>XI</i>	C <sub>13</sub> H <sub>8</sub> ClNO <sub>2</sub> <sup>a</sup> (245·66)	63·55	3·28	5·70	197·5–199 (65·3)	ethanol
		63·62	3·33	5·77		
<i>XII</i>	C <sub>13</sub> H <sub>8</sub> BrNO <sub>2</sub> <sup>b</sup> (290·11)	53·82	2·78	4·83	177–179 (65·3)	ethanol
		53·84	2·72	4·89		
<i>XIII</i>	C <sub>13</sub> H <sub>8</sub> INO <sub>2</sub> (337·11)	46·31	2·39	4·15	130(decomp.) (95·6)	ethanol
		46·49	2·52	4·21		
<i>XIV</i>	C <sub>14</sub> H <sub>11</sub> NO <sub>2</sub> (225·24)	74·65	4·92	6·62	144–145·5 (73·9)	ether–light petroleum
		74·84	4·99	6·36		
<i>XV</i>	C <sub>14</sub> H <sub>11</sub> NO <sub>2</sub> S <sup>c</sup> (257·30)	65·35	4·31	5·44	180(decomp.) (81·7)	ether–light petroleum
		65·42	4·47	5·40		
<i>XVI</i>	C <sub>14</sub> H <sub>9</sub> NO <sub>4</sub> (255·22)	65·84	3·55	5·48	181–182 (68·0)	ethanol
		65·78	3·70	5·31		
<i>XVII</i>	C <sub>13</sub> H <sub>9</sub> NOS <sup>d</sup> (227·28)	68·69	3·99	6·16	194·5–196 (63·3)	ethanol
		68·72	3·88	6·13		
<i>XVIII</i>	C <sub>13</sub> H <sub>8</sub> N <sub>2</sub> O <sub>3</sub> S <sup>e</sup> (272·28)	57·34	2·96	10·29	261–263 (72·1)	ethanol
		57·45	3·04	10·32		

<sup>a</sup> Calculated 14·14% Cl, found 14·41% Cl; <sup>b</sup> calculated 27·54% Br, found 27·84% Br; <sup>c</sup> calculated 12·44% S, found 12·46% S; <sup>d</sup> calculated 14·11% S, found 14·21% S; <sup>e</sup> calculated 11·77% S, found 11·74% S; compounds *IX*, *X*, *XVII*, *XVIII* were prepared by Method *A* and compounds *IX*, *XI–XVI* by Method *B*.

to  $\nu(\text{C}=\text{O})$  of the cycle and the band of lower frequency ( $1688-1665\text{ cm}^{-1}$ ) to  $\nu(\text{C}=\text{O})$  of the  $\text{COCH}_3$  group (see ref.<sup>16</sup>). The assignment of other distinct bands is the following:  $\nu(\text{C}=\text{C})_{\text{alif.}}$   $1630\text{ cm}^{-1}$ ,  $\nu(\text{C}=\text{C})_{\text{aromat.}}$   $1622-1575\text{ cm}^{-1}$  and  $1500\text{ cm}^{-1}$  region,  $\nu(\text{C}_{\text{Ar}}-\text{N})$   $1350\text{ cm}^{-1}$  region,  $\nu(\text{C}-\text{O}-\text{C})_{\text{as}}$   $1260-1230\text{ cm}^{-1}$ ,  $\nu(\text{C}-\text{O}-\text{C})_{\text{s}}$   $1083-1011\text{ cm}^{-1}$ . In compounds *I-VIII*  $\delta$  sym  $(\text{CH}_3)_{\text{COCH}_3}$  was

TABLE III

IR (KBr) and UV (ethanol) Spectral Parameters of Substituted 2-(2-Furfurylidene- and 2-Thienylidene)indoxyls *I-XVIII*;  $\tilde{\nu}(\text{cm}^{-1})$ ;  $\lambda_{\text{max}}$  (nm)

Compound	$\nu(\text{C}=\text{O})$	$\nu(\text{C}=\text{C})_{\text{alif}}$	$\nu(\text{C}-\text{O}-\text{C})_{\text{s}}$	$\lambda_{\text{max}}$ (log $\epsilon$ )			
<i>I</i>	1 693 1 676	1 628	1 022	204 (4·35)	257 (4·19)	347 (4·21)	431 (4·09)
<i>II</i>	1 709 1 684	1 619	1 030	225 (4·19)	259 (4·21)	353 (4·34)	442 (4·16)
<i>III</i>	1 698 1 684	1 610	1 028	223 (4·03)	268 (4·06)	357 (3·96)	446 (4·16)
<i>IV</i>	1 692 1 685	1 618	1 027	228 (4·12)	276 (4·11)	370 (3·93)	456 (4·27)
<i>V</i>	1 716 1 688	1 622	1 028	215 (4·04)	254 (4·04)	337 (4·30)	429 (3·86) 528 (3·55)
<i>VI</i>	1 722 1 681	1 608	1 011	222 (4·07)	262 (4·09)	340 (4·50)	440 (3·72)
<i>VII</i>	1 689 1 665	1 627	825 <sup>a</sup>	213 (4·36)	266 (4·21)	341 (4·20)	438 (4·12)
<i>VIII</i>	1 702 1 672	1 616	820 <sup>a</sup>	213 (4·12)	259 (4·21)	261 (4·25)	454 (4·09)
<i>IX</i>	1 693	1 631	1 038	233 (4·18)	296 (4·18)	364 (4·07)	495 (4·16)
<i>X</i>	1 704	1 631	1 028	232 (4·13)	274 (4·41)	373 (4·18)	572 (4·27)
<i>XI</i>	1 695	1 630	1 032	236 (4·25)	303 (4·19)	367 (4·10)	492 (4·21)
<i>XII</i>	1 688	1 627	1 031	236 (4·28)	303 (4·23)	369 (4·15)	500 (4·27)
<i>XIII</i>	1 679	1 621	1 029	237 (4·17)	300 (4·09)	373 (4·05)	500 (4·20)
<i>XIV</i>	1 685	1 630	1 048	235 (4·20)	299 (4·06)	375 (4·05)	500 (4·21)
<i>XV</i>	1 688	1 631	1 030	241 (4·18)	304 (4·05)	387 (3·97)	682 (4·28)
<i>XVI</i>	1 719 1 687	1 638	1 031	246 (4·33)	330 (4·20)	373 (3·90)	681 (4·15)
<i>XVII</i>	1 691	1 634	858 <sup>a</sup>	237 (4·08)	313 (4·11)	361 (4·03)	490 (4·10)
<i>XVIII</i>	1 700	1 633	841 <sup>a</sup>	234 (4·15)	276 (6·33)	380 (4·23)	523 (4·33)

<sup>a</sup> Skeletal vibrations of thiophene.

about  $1370\text{ cm}^{-1}$ . The sharp band of  $\nu(\text{N}-\text{H})$  at  $3455\text{ cm}^{-1}$  could not be observed in the case of compounds *IX*–*XVIII* when the spectra were measured in KBr; it was observed in chloroform and tetrachloromethane solutions only<sup>17,18</sup>. Electronic absorption spectra of the investigated substances *I*–*XVIII* display 4 absorption maxima (Table III).

The mass spectrum of compound *X* is shown in Fig. 1. The molecular ion, which is at the same time the most abundant one, indicates the minimum mass of a compound of empirical formula  $\text{C}_{13}\text{H}_8\text{N}_2\text{O}_4$ . The presence of the nitro group is indicated by the fragment ion  $m/e$  210 which is formed after the splitting off of the radical  $\text{NO}_2^{\cdot}$  from the molecular ion. In agreement with the known fragmentation of the nitro group<sup>19</sup> weakly intensive ions ( $\text{M}^{+\cdot} - \text{NO}^{\cdot}$ ),  $m/e$  226, and ( $\text{M}^{+\cdot} - \text{NO}^{\cdot} - \text{CO}$ ),

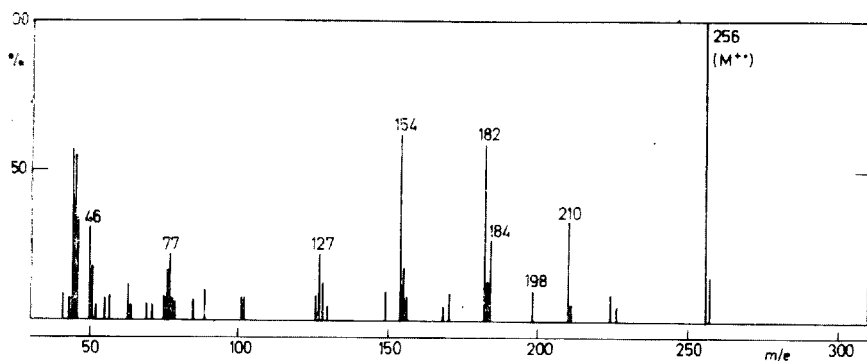


FIG. 1  
Mass Spectrum of 2-(5-Nitro-2-furfurylidene)indoxyl (*X*)

$m/e$  198, were also observed in the spectrum. Further fragments of  $m/e$  182 ( $\text{M}^{+\cdot} - \text{NO}_2 - \text{CO}$ ),  $m/e$  154 ( $\text{M}^{+\cdot} - \text{NO}_2 - \text{CO} - \text{CO}$ ), and  $m/e$  127 ( $\text{M}^{+\cdot} - \text{NO}_2 - \text{CO} - \text{CO} - \text{HCN}$ ) are connected with the cleavage of both heterocyclic nuclei<sup>20,21</sup>. The ion of the fragment  $m/e$  155 is formed by splitting off of HCN from the fragment ( $\text{M}^{+\cdot} - \text{NO}_2 - \text{CO}$ ). The correctness of the mentioned fragmentation is confirmed by the presence of the following metastable ions:  $m^* = 172.3$ , confirming the fragmentation transition of ion ( $\text{M}^{+\cdot}$ ),  $m/e$  256  $\rightarrow$  ( $\text{M}^{+\cdot} - \text{NO}_2^{\cdot}$ ),  $m/e$  210;  $m^* = 157.7$  the transition ( $\text{M}^{+\cdot} - \text{NO}_2^{\cdot}$ ),  $m/e$  210  $\rightarrow$  ( $\text{M}^{+\cdot} - \text{NO}_2 - \text{CO}$ ),  $m/e$  182;  $m^* = 132.0$  the transition ( $\text{M}^{+\cdot} - \text{NO}_2 - \text{CO}$ ),  $m/e$  182  $\rightarrow$  ( $\text{M}^{+\cdot} - \text{NO}_2 - \text{CO} - \text{HCN}$ ),  $m/e$  155.

However, the sign of the metastable ion  $m^* = 104.7$  cannot confirm the fragmentation ( $\text{M}^{+\cdot} - \text{NO}_2 - \text{CO} - \text{CO}$ ),  $m/e$  154  $\rightarrow$  ( $\text{M}^{+\cdot} - \text{NO}_2 - \text{CO} - \text{CO} - \text{HCN}$ ),  $m/e$  127. Further, more important fragment ions of the spectrum,  $m/e$  77 ( $\text{C}_6\text{H}_5$ ), 76 ( $\text{C}_6\text{H}_4$ ), 63 ( $\text{C}_5\text{H}_3$ ), 51 ( $\text{C}_4\text{H}_5$ ), are connected with the presence of both heterocyclic rings and

the nitro group ( $m/e$  45) in the compound investigated. The fragmentation which was observed confirms the correctness of the structures proposed for substituted 2-(2-furfurylidene- or 2-thenylidene)indoxyls.

### EXPERIMENTAL

Melting points were determined on a micromelting point apparatus Boëtius. Samples for analysis were dried at 40°C and 15 Torr for 5 hours. IR spectra were measured on a two-beam prism spectrophotometer UR-20, Zeiss, Jena. Electronic spectra were taken on a registration spectrophotometer Specord UR VIS, Zeiss, Jena. Mass spectra were recorded on a AEI MS 902-S instrument. For the control of purity of final products on Silufol thin-layer plates the following solvent mixtures were employed: benzene-ethyl acetate 4 : 5 and 5 : 1, cyclohexane-ethyl acetate 4 : 1, and methanol-acetone-benzene 1 : 1 : 4 and 1 : 0.5 : 5.

#### 1-Acetyl-2-(5-X-2-furfurylidene or 2-thenylidene)indoxyls I—VIII

0.01 mol of the corresponding 5-substituted 2-furan- or 2-thiophenecarbaldehyde and 4 drops of piperidine were added to a boiling solution of 0.01 mol of 1-acetylindoxyl in 200 ml of benzene and the mixture was stirred and refluxed for 2 hours. Charcoal was then added and the mixture filtered and the filtrate evaporated under reduced pressure. Compounds I—VIII were crystallized from a suitable solvent. Compound II was prepared by condensation at room temperature. In the case of compound VI the reaction time was extended to 4 hours.

#### 2-(5-X-2-Furfurylidene or 2-thenylidene)indoxyls IX, X, XVII, XVIII

Dilute hydrochloric acid (1 : 1; 3.3 ml) was added dropwise over 45 minutes to a boiling solution of 0.01 mol of 1-acetyl-2-(5-X-2-furfurylidene or 2-thenylidene)indoxyl (I, II, VII, VIII) in 120 ml of ethanol and the reaction mixture was refluxed for a further 45 minutes. After dilution with water 90 ml of aqueous ethanol were eliminated from it by distillation. After the repeated addition of 50 ml of water, cooling and alkalization with aqueous ammonia to pH 7.0—7.5 the crude reaction product precipitated gradually. It was purified by crystallization. For hydrolysis of the poorer soluble compound XVIII acetone or propanol were employed.

#### 2-(5-X-2-Furfurylidene)indoxyls IX, XI—XVI

In a closed flask a mixture of 0.016 mol of 3-acetylindoxyl, 0.006 mol of the corresponding carbonyl compound, 50 ml of water, and 3.3 ml of 4M-KOH were heated at 30—40°C under occasional shaking. A coloured precipitate began to form immediately. The end of the reaction was indicated by the disappearance of the starting aldehyde (approximately after 2 hours). The product was extracted with ether, the extract dried over anhydrous sodium sulfate, filtered with charcoal and evaporated to a small volume under reduced pressure. After cooling a solid product was obtained which was purified by crystallization. The working procedure was modified for the synthesis of compound XVI. From the alkali salt of the acid formed by hydrolysis of its ester free acid is obtained by acidification of the reaction mixture with conc. HCl.

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