NEW 4-SUBSTITUTED 1,2-DIPHENYL-3,5-DIOXOPYRAZOLIDINES

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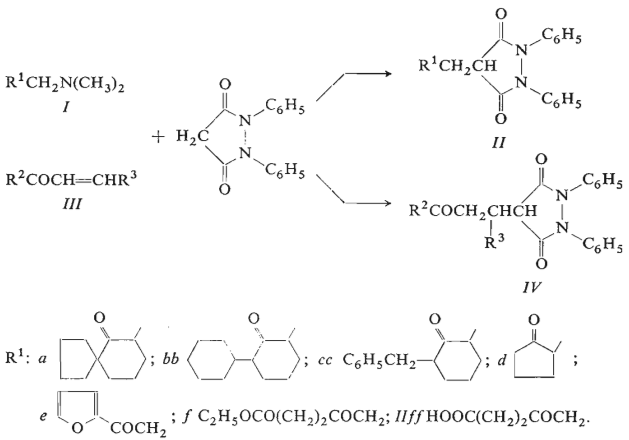
Novel 4-substituted derivatives were prepared from the sodium salt of 1,2-diphenyl-3,5-dioxopyrazolidine by alkylation with Mannich bases and addition to α,β-unsaturated ketones. The carboxymethyl and dimethylaminoethyl groups were introduced by alkylation of the sodium salts into position 4 of some 4-monosubstituted derivatives. Fischer's reaction of some keto derivatives of 1,2-diphenyl-3,5-dioxopyrazolidine with N-(4-chlorobenzoyl)-N-(4-methoxy-phenyl) hydrazine led to the corresponding indole derivatives.

The present work was stimulated by the fact that several years after practical application of the anti-inflammatory agent 1,2-diphenyl-3,5-dioxo-4-butylpyrazolidine[1] (phenylbutazone) two highly active compounds in the same area, *viz.* 1,2-diphenyl-3,5-dioxo-4-(3-oxobutyl)pyrazolidine[2,3] (ketophenylbutazone) and 1,2-diphenyl-3,5-dioxo-4-(4,4-dimethyl-3-oxopentyl)pyrazolidine[4] (trimetazone) have been prepared in this Institute. In this connection several new compounds have been prepared[5] with a view to the various structural possibilities given by the presence of one or two active hydrogen atoms in position 4 of the 1,2-diphenyl-3,5-dioxopyrazolidine molecule.

The compounds mentioned here are divided into four groups. The first of these is formed by the derivatives of 1,2-diphenyl-3,5-dioxopyrazolidine containing a side chain in position 4, the position 3 bearing a carbonyl group (*IIa*–*IIf*, *IIff*). Their preparation consists in an alkylation of the active methylene group of the dioxopyrazolidine ring with ketonic Mannich bases (*I*). Use is made here of the finding of a positive effect of an equivalent amount of alcoholate[2–4] for the reaction. The intermediate is thus the sodium salt of dioxopyrazolidine which is added in the form of a carbanion to the double bond of the vinyl ketone formed from the Mannich base. An analogous addition of dioxopyrazolidine to α,β-unsaturated ketones yielded compounds of the second group (*IVa*–*IVl*). Generally, the required pyrazolidines of the first and second groups can be obtained by a reaction of a freshly prepared sodium salt of 1,2-diphenyl-3,5-dioxopyrazolidine with ketonic Mannich base or with α,β-unsaturated ketone in dimethylformamide. The acids of both groups were obtained by alkaline hydrolysis of the esters. When preparing *IIc* it was attempted to introduce the benzylidene group into position 3 of the cyclohexanone

ring of 1,2-diphenyl-3,5-dioxo-4-(2-oxo-1-cyclohexyl)methylpyrazolidine[5] with the aim of reducing the double bond but with no success.



R¹: *a* ; *bb* ; *cc* $C_6H_5CH_2$- ; *d* ;

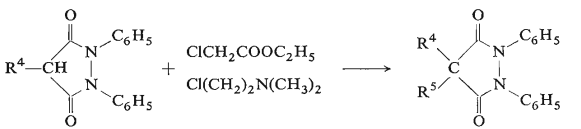
e ; *f* $C_2H_5OCO(CH_2)_2COCH_2$; *IIff* $HOOC(CH_2)_2COCH_2$.

R^2, R^3: *a* CH_3, CH_3; *b* C_6H_5, 4-$NO_2C_6H_4$; *c* C_6H_5, 4-$(CH_3)_2NC_6H_4$; *d* 4-pyridyl, C_6H_5; *e* 5-indanyl, C_6H_5; *f* 2-furyl, C_6H_5; *g* 2-furyl, 2-furyl; *h* 4-$(CH_3)_3CC_6H_4$, 2-furyl; *i* CH_3, $COOC_2H_5$; *j* $(CH_3)_3C$, $COOC_2H_5$; *k* C_6H_5, $COOCH_3$; *l* 1-adamantyl, $COOC_2H_5$; *IVii*–*IVll* R^3: COOH.

Mannich bases or their hydrochlorides of compounds of the first group were prepared by described procedures[6] involving the reaction of saturated or unsaturated ketones with formaldehyde or paraformaldehyde and secondary amine. The double bond of the hydrochlorides of unsaturated bases was hydrogenated on palladium[7]. With the exception of the derivative of cyclopentanone and laevulinic acid, all the bases were used after liberation from their hydrochlorides in a crude state. Of the Mannich bases, derivatives *Ia*–*Ic* and *Ibb*, *Icc* are new. To prepare α,β-unsaturated ketones the method common in the chemistry of chalcones was used[8]. With the exception of 1-(2-furyl)-3-(4-t-butylphenyl)-3-oxopropene (*IIIh*) all have been described before. Acetyl, pivaloyl and 1-adamantoylacrylic acids or their esters were prepared by Wittig's reaction[9]. Benzoylacrylic acid was obtained by the Friedel–Crafts reaction[10] and converted to its ester by hydrogen chloride in methanol.

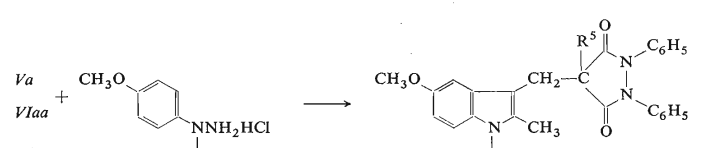
The starting compound for compounds of the third group (*VIa*–*VIg*) were some pharmacologically more important 4-substituted derivatives of dioxopyrazolidine

(*Va*–*Vd*) where the remaining atom of active hydrogen was replaced with a carboxymethyl or a dimethylaminoethyl group in the form of hydrochloride. In this category of compounds, changes in pharmacological properties brought about by replacing the active hydrogen atom with another acidic function or with a substituent that will increase the water solubility of the compound were examined. The method of preparation here was alkylation of the sodium salt of dioxopyrazolidine derivatives *V* with ethyl chloroacetate in dimethylformamide (*VIa*–*VIc*) and dimethylaminoethyl chloride in toluene (*VId*–*VIg*). Derivatives of acetic acid esters of this group of compounds (*VIa*–*VIc*) were prepared by a somewhat modified procedure applied for the analogous ester of phenylbutazone[11]. In attempts at obtaining acids *VIaa* to *VIcc* from esters it was observed that alkaline hydrolysis can be used only for the phenylbutazone derivative. With all the other esters containing a carbonyl group in position 3 of the side chain of the dioxopyrazolidine ring, a retro-Michael addition took place under conditions of alkaline hydrolysis, resulting in splitting off the side chain. The reaction products here were 1,2-diphenyl-3,5-dioxo-4-carboxymethylpyrazolidine and the corresponding vinylketone. All the acids of this group were obtained by hydrolysis carried out in a mixture of acetic and hydrochloric acids. The dimethylaminoethylpyrazolidines *VId*–*VIg* were converted to the hydrochlorides without prior isolation. (All the hydrochlorides prepared exhibited the expected water solubility.)



V *VI*

R^4: *a*, *d* $CH_3CO(CH_2)_2$; *b*, *e* $C_6H_5CO(CH_2)_2$; *c*, *f* $(CH_3)_3CCO(CH_2)_2$; *g* $CH_3(CH_2)_3$.
R^5: *a*–*c* $C_2H_5OCOCH_2$; *aa*–*cc* $HOOCCH_2$; *d* $(CH_3)_2N(CH_2)_2$; *e*–*g* $HClN(CH_3)_2(CH_2)_2$.



VIIa, $R^5 = H$
VIIb, $R^5 = CH_2COOH$

The series of compounds prepared here is concluded by two derivatives which, by their structure and method of preparation, stand apart from the above three groups but which can be compared with phenylbutazone. Their molecules contained an indole ring which was built up by Fischer's reaction. The starting components were N-(4-chlorobenzoyl)-N-(4-methoxyphenyl)hydrazine hydrochloride[12,13], ketophenylbutazone (*Va*) and 4-carboxymethylketophenylbutazone (*VIaa*). The corresponding N-acyl-N-phenylhydrazine was prepared by direct acylation of 4-methoxyphenylhydrazine hydrochloride. This part of our work proceeded from earlier findings connected with the synthesis of indomethacine (1-(4-chlorobenzoyl)-2-methyl-5-methoxy-3-indolylacetic acid).

Of all the compounds reported here the most interesting pharmacologically was found to be the second group, in particular the derivatives of dioxopyrazolidine bearing a carboxyl group at the α-carbon of their side chain.

EXPERIMENTAL

The melting points are not corrected. The IR spectra were measured on an Infrascan Hilger spectrophotometer in the form of KBr pellets.

Hydrochlorides of Mannich Bases *Ia*—*Ic* (Table I)

A mixture of 0·1 mol ketone, 0·1 mol dimethylamine hydrochloride and 0·13 mol paraformaldehyde was boiled in 17 ml 95% ethanol with 0·25 ml concentrated hydrochloride acid for 6 h (*Ia*), for 3 h (*Ib*) and for 1 h (*Ic*). After cooling, the precipitated crystalline product was mixed with acetone, filtered and recrystallized. The starting compound for *Ia* was 6-oxospiro[4,5]decane[14], for *Ib* cyclohexylidenecyclohexanone[15] and for *Ic* benzylidenecyclohexanone[16].

Hydrogenation of *Ib*, *Ic* to *Ibb*, *Icc* (Table I)

0·063 mol *Ib* was hydrogenated on 4 g Pd/C in 500 ml ethanol to *Ibb*; 0·034 mol *Ic* was hydrogenated on 2 g Pd/C in 350 ml water to *Icc*. After filtration of the catalyst, the solvent was distilled at reduced pressure to dryness, the residue was mixed with acetone, the crystalline compound was filtered and recrystallized.

Alkylation of 1,2-Diphenyl-3,5-dioxopyrazolidine by Mannich Bases (*IIa*—*IIf*, Table II)

A solution of 0·1 mol sodium methylate in methanol was added to 0·1 mol 1,2-diphenyl-3,5-dioxopyrazolidine in methanol and the solvent was evaporated to dryness at reduced pressure. 200 ml dimethylformamide and 0·1 mol of the corresponding Mannich base was added to the sodium salt thus prepared. The Mannich bases used were: 1-dimethylaminomethyl-2-oxocyclopentane[17] (*Id*) for *IId*, 1-(2-furyl)-1-oxo-3-dimethylaminopropane[18] (*Ie*) for *IIe*, ethyl ester of 6-piperidino-4-oxocaproic acid[19] (*If*) for *IIf*. The reaction mixture was heated for 3 h to 115—120°C. After cooling, it was poured into 500 ml water and acidified with dilute hydrochloric acid (1 : 1). The precipitated product was filtered, washed with water and recrystallized.

Ie and *If* were purified by distillation. The Mannich bases of the spiroketone *Ia*, cyclohexylcyclohexanone *Ibb* and benzylcyclohexanone *Icc* were liberated from the methanolic solution

TABLE I

Hydrochlorides of Mannich Bases *I*

I (yield, %)	M.p., °C (solvent)	Formula (M.w.)	Calculated/Found			
			% C	% H	% Cl	% N
Ia (40)	138—140 (methylethyl ketone)	$C_{13}H_{24}ClNO$ (245·8)	63·40 63·56	9·80 10·00	14·42 14·28	5·7 5·73
Ib (42)	157—159 (ethanol)	$C_{15}H_{26}ClNO$ (271·8)	66·26 66·23	9·64 9·78	13·04 13·10	5·15 5·10
Ibb (85)	161—162 (2-propanol)	$C_{15}H_{28}ClNO$ (273·8)	65·78 65·60	10·30 10·34	13·15 12·90	5·11 5·30
Ic (52)	150—151 (ethanol)	$C_{16}H_{22}ClNO$ (279·8)	68·69 68·55	7·89 7·95	12·67 12·66	5·07 4·87
Icc (84)	156—157 (2-propanol)	$C_{16}H_{24}ClNO$ (281·8)	68·20 68·00	8·58 8·63	12·58 12·60	4·97 4·92

TABLE II

Pyrazolidines *II*

II (yield, %)	M.p., °C (solvent)	Formula (M.w.)	Calculated/Found		
			% C	% H	% N
IIa (58)	160—162 (ethanol)	$C_{26}H_{28}N_2O_3$ (416·5)	74·97 75·01	6·78 6·81	6·73 6·76
IIb (55)	177—179 (2-propanol)	$C_{28}H_{32}N_2O_3$ (444·6)	75·65 75·70	7·26 7·15	6·30 6·15
IIc (40)	138—140 (ethanol)	$C_{29}H_{28}N_2O_3$ (452·5)	76·97 76·96	6·24 6·32	6·19 6·39
IId (59)	148—150 (ethanol)	$C_{21}H_{20}N_2O_3$ (348·4)	72·39 72·53	5·79 5·99	8·04 8·28
IIe (58)	142—143 (ethanol)	$C_{22}H_{18}N_2O_4$ (386·4)	70·58 70·36	4·85 4·90	7·48 7·24
IIf (62)	95—96 (ethanol)	$C_{23}H_{24}N_2O_5$ (408·4)	67·63 67·72	5·92 5·81	6·86 7·07
IIff (64)	148—150 (benzene)	$C_{21}H_{20}N_2O_5$ (380·4)	66·30 66·15	5·30 5·24	7·37 7·31

of the hydrochlorides with an equivalent amount of sodium methylate in methanol and, after removing the solvent, used in the form of residue after evaporation. With the furane derivative *Ie*, the base was liberated from the solution of hydrochloride in dimethylformamide (0·1 mol in 50 ml) by a suspension of sodium methylate in dimethylformamide (0·1 mol in 50 ml) and the salt solution was added to the solution of dioxopyrazolidine.

Hydrolysis of Ester *IIf* to Acid *IIff* (Table II)

0·05 mol ester *IIf* was boiled for 2 h in a mixture of 200 ml 3% sodium hydroxide and 400 ml ethanol. After cooling, the mixture was acidified with dilute hydrochloric acid (1 : 1) and the product was extracted with dichloromethane. The solution was washed with water, concentrated to dryness and the crude acid thus isolated.

1-(2-Furyl)-3-(4-t-butylphenyl)-3-oxopropene (*IIIh*)

Using the procedure for the preparation of benzalacetophenone[8], an unsaturated ketone was prepared from 0·2 mol 4-t-butylacetophenone and 0·2 mol furfural, in a 53% yield, m.p. 80 to 82°C (ethanol). For $C_{17}H_{18}O_2$ (254·3) calculated: 80·28% C, 7·13% H; found: 80·11% C, 7·12% H.

Addition of 1,2-Diphenyl-3,5-dioxopyrazolidine to α,β-Unsaturated Ketones (*IVa—IVl*, Table III)

A mixture of 0·1 mol sodium salt 1,2-diphenyl-3,5-dioxopyrazolidine prepared by the procedure described for alkylation with Mannich bases, and of 0·1 mol ketone was heated for 3 h in 200 ml dimethylformamide to 100°C. The method of isolation of these compounds was identical with the procedure described for alkylation. During addition of dioxopyrazolidine to the ester of acetylacrylic acid, the ester formed was hydrolyzed simultaneously and the desired acid *IVii* was isolated.

Ketones used: 2-oxo-3-pentene[20] (*IIIa*) for *IVa*, 1-(4-nitrophenyl)-3-phenyl-3-oxopropene[21] (*IIIb*) for *IVb*, 1-(4-dimethylaminophenyl)-3-phenyl-3-oxopropene[22] (*IIIc*) for *IVc*, 1-phenyl-3-(4-pyridyl)-3-oxopropene[23] (*IIId*) for *IVd*, 1-phenyl-3-(5-indanyl)-3-oxopropene[24] (*IIIe*) for *IVe*, 1-phenyl-3-(2-furyl)-3-oxopropene[25] (*IIIf*) for *IVf*, 1-(2-furyl)-3-(2-furyl)-3-oxopropene[25] (*IIIg*) for *IVg*, 1-(2-furyl)-3-(4-tert-butylphenyl)-3-oxopropene (*IIIh*) for *IVh*, ethyl esters of acetyl, pivaloyl and 1-adamantoylacrylic acids *IIIi*, *IIIj*, *IIIl* (ref.[9]) for *IVi*, *IVj*, *IVl*, benzoylacrylic acid[10] and its methyl ester *IIIk* for *IVk*. (The acid was esterified with methanolic hydrogen chloride, b.p. of the ester 165—170°C/12 Torr, 191°C/40 Torr (ref.[26])).

Hydrolysis of Esters *IVj—IVl* to Acids *IVjj—IVll* (Table III)

The esters were hydrolyzed in the corresponding alcohols in the same way as described for the ester *IIf*. The yields given in Table III refer to the starting 1,2-diphenyl-3,5-dioxopyrazolidine.

Preparation of Esters *VIa—VIc* (Table IV)

A mixture of 0·044 mol sodium salt of pyrazolidine *V* monosubstituted in position 4, prepared as described for the alkylation with Mannich bases, and 0·05 mol ethyl chloroacetate was heated for 5 h in 80 ml dimethylformamide to 100°C. After distillation of the solvent at reduced pressure to dryness the residue was mixed with water, the crystalline product was filtered and recrystallized. 1,2-Diphenyl-3,5-dioxopyrazolidines substituted in position 4 were used: with

TABLE III
Pyrazolidines *IV*

IV (yield, %)	M.p., °C (solvent)	Formula (M.w.)	Calculated/Found		
			% C	% H	% N
IVa (65)	127—128 (2-propanol)	$C_{20}H_{20}N_2O_3$ (336·4)	71·41 71·51	5·99 6·09	8·33 8·17
IVb (45)	157—158 (methylcellosolve)	$C_{30}H_{23}N_3O_5$ (505·5)	71·27 70·95	4·59 4·67	8·31 8·07
IVc (40)	159—160 (benzene–cyclo--hexane 1 : 1)	$C_{32}H_{29}N_3O_3$ (503·6)	76·32 76·47	5·80 5·75	8·35 8·13
IVd (38)	187—188 (ethanol)	$C_{29}H_{23}N_3O_3$ (461·5)	75·47 75·33	5·02 5·13	9·11 8·94
IVe (64)	165—167 (ethanol)	$C_{33}H_{28}N_2O_3$ (500·6)	79·18 78·90	5·64 5·71	5·60 5·79
IVf (70)	158—160 (ethanol)	$C_{28}H_{22}N_2O_4$ (450·5)	74·65 74·64	4·92 5·06	6·22 6·25
IVg (76)	159—160 (methylcellosolve)	$C_{26}H_{20}N_2O_5$ (440·4)	70·90 70·73	4·58 4·69	6·36 6·29
IVh (65)	134—135 (ethanol)	$C_{32}H_{30}N_2O_4$ (506·6)	75·87 75·81	5·97 6·02	5·53 5·74
IVii (52)	194—195 (2-propanol)	$C_{20}H_{18}N_2O_5$ (366·4)	65·56 65·76	4·95 5·00	7·65 7·75
IVj (80)	115—116 (2-propanol)	$C_{25}H_{28}N_2O_5$ (436·5)	68·79 69·00	6·47 6·47	6·42 6·20
IVjj (45)	180—182 (benzene–cyclo-hexane 1 : 2)	$C_{23}H_{24}N_2O_5$ (408·4)	67·63 67·90	5·92 6·08	6·86 7·71
IVk (53)	135—136 (2-propanol)	$C_{26}H_{22}N_2O_5$ (442·5)	70·58 70·77	5·01 4·93	6·33 6·16
IVkk (26)	190—191 (ethanol–water 2 : 1)	$C_{25}H_{20}N_2O_5$ (428·4)	70·08 69·91	4·71 4·77	6·54 6·63
IVl (76)	152—153 (ethanol)	$C_{31}H_{34}N_2O_5$ (514·6)	72·35 72·15	6·66 6·70	5·44 6·64
IVll (48)	224—225 (2-propanol)	$C_{29}H_{30}N_2O_5$ (486·6)	71·58 71·47	6·22 6·17	5·76 5·55

3-oxobutyl[2,3] (*Va*) for *VIa*, with 3-phenyl-3-oxopropyl[5] (*Vb*) for *VIb*, with 4,4-dimethyl-3-oxopentyl[4] (*Vc*) for *VIc*.

Hydrolysis of *VIa*—*VIc* to Acids *VIaa*—*VIcc* (Table IV)

A mixture of 0·04 mol ester, 270 ml acetic acid and 135 ml concentrated hydrochloric acid was left for 3 days at 35—40°C. After dilution of the mixture with 3000 ml water, the acid was extracted with ether from a solution saturated with sodium chloride. Crystallization of the residue of the ether solution yielded the desired product. The yields of Table IV refer to the starting 4-monosubstituted dioxopyrazolidine *V*.

TABLE IV
Pyrazolidine *VI*

VI (yield, %)	M.p., °C (solvent)	Formula (M.w.)	Calculated/Found		
			% C	% H	% N
VIa (65)	117—118 (2-propanol)	$C_{23}H_{24}N_2O_5$ (408·4)	67·63 67·35	5·92 5·73	6·86 7·08
VIaa (67)	170—171 (2-propanol)	$C_{21}H_{20}N_2O_5$ (380·4)	66·30 66·15	5·30 5·43	7·37 7·04
VIb (52)	123—124 (benzene–cyclohexane 1 : 2)	$C_{28}H_{26}N_2O_5$ (470·5)	71·47 71·39	5·57 5·34	5·95 5·87
VIbb (46)	184—185 (ethanol–water 2 : 1)	$C_{26}H_{22}N_2O_5$ (442·5)	70·58 70·30	5·01 5·30	6·33 6·24
VIc (72)	151—153 (ethanol)	$C_{26}H_{30}N_2O_5$ (450·5)	69·31 68·98	6·71 6·54	6·22 6·39
VIcc (50)	176—177 (ethanol–water 3 : 2)	$C_{24}H_{26}N_2O_5$ (422·5)	68·23 68·48	6·20 6·24	6·63 6·58
VId (20)	104—105 (cyclohexane)	$C_{23}H_{27}N_3O_3$ (393·5)	70·20 70·53	6·92 6·78	10·68 10·63
VIe[a] (40)	240—242 (ethanol)	$C_{28}H_{30}ClN_3O_3$ (492·0)	68·35 68·38	6·14 5·83	8·54 8·72
VIf[a] (53)	226—227 (2-propanol)	$C_{26}H_{34}ClN_3O_3$ (472·0)	66·16 65·92	7·26 7·33	8·93 8·90
VIg[a] (30)	215—216 (2-propanol)	$C_{23}H_{30}ClN_3O_3$ (415·9)	66·41 66·55	7·29 6·97	10·13 9·97

[a] % Cl: calculated/found for *VIe* 7·21/7·25; for *VIf* 7·51/7·44; for *VIg* 8·53/8·49.

Preparation of Amines *VId—VIg* (Table IV)

A solution of 10 g potassium hydroxide in 10 ml water was added dropwise at 5°C to a solution of 20 g dimethylaminoethylchloride hydrochloride in a minimal amount of water, cooling was interrupted and the mixture was stirred for 10 min. The lower layer containing the base was dried with potassium hydroxide and, at 40—43°C/58—60 Torr redistilled (9·5 g, 64%). 0·044 mol sodium salt of pyrazolidine *V* substituted in position 4, prepared as described for alkylation with Mannich bases, was heated for 6 h with 0·088 mol dimethylaminoethylchloride in 150 ml toluene to 120°C. After cooling, the precipitated sodium chloride was filtered and the solution concentrated at reduced pressure to dryness. The residue was dissolved in 250 ml anhydrous ether and, under stirring at 5°C, 15 ml anhydrous ether with 0·05 mol hydrogen chloride was added to the ether solution of the base. The mixture was left to stand for 24 h at room temperature, the crystalline compound was filtered and recrystallised.

Since attempts at purification of *VId* were not successful, the base was liberated from the hydrochloride (its content by titration was 96%) with 40% potassium hydroxide and the product was purified in this form.

The following 1,2-diphenyl-3,5-dioxopyrazolidines substituted in position 4 were used: with 3-oxobutyl[2,3] (*Va*) for *VId*, with 3-phenyl-3-oxopropyl[5] (*Vb*) for *VIe*, with 4,4-dimethyl-3-oxopentyl[4] (*Vc*) for *VIf*, with butyl[1] (*Vd*) for *VIg*.

1,2-Diphenyl-3,5-dioxo-4-(1-*p*-chlorobenzoyl-2-methyl-5-methoxy-3-indolylmethyl)pyrazolidine (*VIIa*) and Its Carboxymethyl Derivative *VIIb*

0·6 ml 100% phosphoric acid and 0·02 mol *Va* or *VIaa* was added under stirring at 70°C to a suspension of 0·02 mol N-(4-chlorobenzoyl)-N-(4-methoxyphenyl)-hydrazine[12] in 65 ml anhydrous toluene. The mixture was heated for 1·5 h (*VIIa*) or for 3 h (*VIIb*) to 78—80°C. After cooling, the precipitate was filtered, washed with water and recrystallized. Yield of *VIIa* was 56%, m.p. 187—188°C (2-propanol). For $C_{33}H_{26}ClN_3O_4$ (564·0) calculated: 70·27% C, 4·65% H, 6·29% Cl, 7·45% N; found: 70·39% C, 4·95% H, 6·20% Cl, 7·37% N. Yield of *VIIb* 60%, m.p. 210—211°C (nitromethane). For $C_{35}H_{28}ClN_3O_6$ (622·1) calculated: 67·57% C, 4·53% H, 5·72% Cl, 6·75% N; found: 67·26% C, 4·69% H, 5·69% Cl, 6·79% N.

The elementary analyses were done at the analytical department of this institute under the direction of Dr J. Körbl.

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