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Reactions with Pyrrolidine-2,4-diones, I. New Synthesis of Pyrano[2,3—c]pyrrole and Pyrrolo[3,4—b]pyridine Systems

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The synthesis of some substituted 4-hydroxy-2,5,6,7-tetrahydro-pyrano[2,3—c]pyrrole-2,5-diones (**5**) and 4-hydroxy-1,2,6,7-tetrahydro-5*H*-pyrrolo[3,4—b]pyridine-2,5-diones (**6**) by reacting 1,5-diaryl-pyrrolidine-2,4-diones (**1**) and 1,5-diaryl-1,5-dihydro-4-amino-2*H*-pyrrol-2-ones (**3**) with bis-2,4,6-trichlorophenyl malonates (**4**) is described.

(Keywords: Bis-2,4,6-trichlorophenyl malonates; 1,5-Diaryl-1,5-dihydro-4-amino-2H-pyrrol-2-ones; 1,5-Diarylpyrrolidine-2,4-diones)

Reaktionen mit Pyrrolidin-2,4-dionen, I.
Eine neue Synthese von Pyrano[2,3—c]pyrrol- und Pyrrolo[3,4—b]pyridin-Systemen

Die Synthese einiger substituierter 4-Hydroxy-2,5,6,7-tetrahydro-pyrano[2,3—c]pyrrol-2,5-dione (**5**) und 4-Hydroxy-1,2,6,7-tetrahydro-5*H*-pyrrolo[3,4—b]pyridin-2,5-dione (**6**) gelingt durch Reaktion von 1,5-Diaryl-pyrrolidin-2,4-dion (**1**) bzw. 1,5-diaryl-1,5-dihydro-4-amino-2*H*-pyrrol-2-on (**3**) mit Malonsäure-bis-2,4,6-trichlorphenylester (**4**).

Introduction

The synthesis and some reactions of 1,5-diphenyl- and 1-(3-methylphenyl)-5-phenylpyrrolidine-2,4-diones (**1 a** and **1 b**, respectively) (sub-

* Dedicated to Prof. Dr. Dr. h. c. *O. Kratky*, Graz, on the occasion of his 80th birthday.

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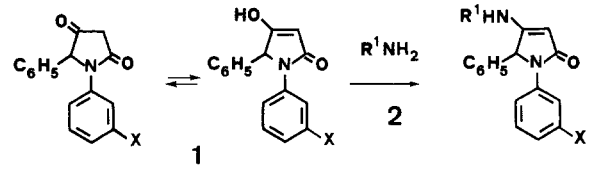
stituted tetramic acids) have been previously investigated by one of us[1]. The reactivity of position 3 of these compounds has been indicated by the ease of formation of 3-arylmethylenepyrrolidine-2,4-diones and by diazo-coupling. Further, the condensation of **1** with primary or secondary amines has afforded the corresponding 1,5-diaryl-1,5-dihydro-4-substituted amino-2*H*-pyrrol-2-ones. In addition, a successful new approach to the synthesis of pyrrolo[3,4—d]-1,2,3-triazole derivatives from **1 a** has been reported[2].

Results and Discussion

Continued interest in the chemistry of pyrrolidine-2,4-diones and their utility for the synthesis of heterocycles with valuable biological properties led us to examine the reaction of **1 a**, **1 b** and some of their 4-substituted amino derivatives (**3**) with bis-2,4,6-trichlorophenyl malonates (**4**). The desired enamines **3** were prepared by condensing **1 a** or **1 b** with the appropriate primary amine **2** in benzene, as illustrated in Scheme 1.

Scheme 1



1	X
a	H
b	CH_3

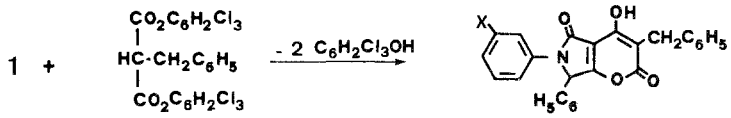
2	R^1
a	C_4H_9
b	$C_6H_5CH_2$
c	C_6H_5

3	R^1	X
a	C_4H_9	CH_3
b	$C_6H_5CH_2$	H
c	$C_6H_5CH_2$	CH_3
d	C_6H_5	H

As expected, reaction of equimolar proportions of **1 a** or **1 b** with bis-2,4,6-trichlorophenyl benzylmalonate (**4 e**) at 160 °C or in refluxing chlorobenzene, respectively, gave the corresponding 6-aryl-3-benzyl-4-hydroxy-7-phenyl-2,5,6,7-tetrahydro-pyrano[2,3—c]pyrrole-2,5-diones (**5 a**, **5 b**) in low yields. Similarly, substituted 4-hydroxy-7-phenyl-1,2,6,7-tetrahydro-5*H*-pyrrolo[3,4—b]pyridine-2,5-diones (**6**, Table 1) were obtained in good yields by fusing **3** with unsubstituted or monosubstituted bis-2,4,6-trichlorophenyl malonates (**4**). The cycliza-

tion involves the interaction between the two 1,3-nucleophilic centers of **1** or **3** with a reactive ketene-carboxylate intermediate[3] which is generated *in situ* from **4** on thermolysis.

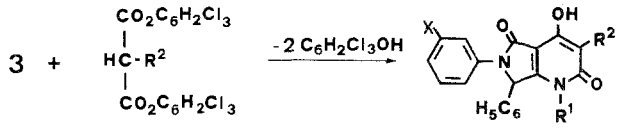
Scheme 2



4e

5	X
a	H
b	CH_3

Scheme 3



4	R^2
a	H
b	C_2H_5
c	$(CH_3)_2CH$
d	C_4H_9
e	$C_6H_5CH_2$
f	C_6H_5

6a-g

R, X - KEY SEE TABLE 1

Only three syntheses of a few examples of pyrano[2,3—c]pyrroles have been previously described. 3-Anilino-6-phenylpyrano[2,3—c]pyrrole-2,5,7(6*H*)-trione has been obtained by treating ethyl 3-hydroxy-2-pyrone-5,6-dicarboxylate in dimethylformamide followed by reacting the intermediate chloride with aniline in glacial acetic acid[4]. Substituted 3,4,6,7-tetrahydro-pyrano[2,3—c]pyrrol-5(2*H*)-ones have been prepared from substituted 2-bromomethyl-3-ethoxycarbonyl-5,6-dihydro-4*H*-pyrans or their thermally cyclized derivatives, furopyrans, and primary or secondary amines[5]. Further, the reaction of 1-aminocyclohexanecarboxylic acid or its derivatives with 5,6-dihydro-4*H*-pyran-2,3-dicarboxylic acid anhydrides afforded the substituted 3,4-dihydro-2*H*-pyran[2,3—c]pyrrole-5,7(6*H*)-diones[6].

Table 1. *Substituted 4-hydroxy-7-phenyl-1,2,6,7-tetrahydro-5H-pyrrolo[3,4—b]pyridine-2,4-diones* (**6**)

Compound	R^1	R^2	X	Yield %	M.p. °C	Recryst. solvent	Molecular formular	Analysis Calcd.	Found
6a	C_4H_9	H	CH_3	70.0	253–254	Ethanol	$C_{24}H_{24}N_2O_3$ (388.5)	C 74.20 H 6.23 N 7.21	73.90 6.20 7.03
6b	$C_6H_5CH_2$	H	H	73.5	242–244	Ethanol	$C_{26}H_{20}N_2O_3$ (408.5)	C 76.45 H 4.95 N 6.86	76.24 5.05 7.09
6c	$C_6H_5CH_2$	C_2H_5	CH_3	63.0	203–205	Ethanol	$C_{29}H_{26}N_2O_3$ (450.5)	C 77.31 H 5.82 N 6.22	77.03 5.69 6.27
6d	$C_6H_5CH_2$	$(CH_3)_2CH$	CH_3	56.5	201–203	Ethanol	$C_{30}H_{28}N_2O_3$ (464.5)	C 77.56 H 6.07 N 6.03	77.84 6.19 5.96
6e	C_6H_5	C_4H_9	H	75.5	235–237	Ethanol	$C_{29}H_{26}N_2O_3$ (450.5)	C 77.31 H 5.82 N 6.22	77.80 5.96 6.20
6f	C_6H_5	$C_6H_5CH_2$	H	76.5	278–280	Acetic acid	$C_{32}H_{24}N_2O_3$ (484.6)	C 79.32 H 4.99 N 5.78	79.60 5.00 5.72
6g	C_6H_5	C_6H_5	H	66.0	312 dec.	Ethanol	$C_{31}H_{22}N_2O_3$ (470.5)	C 79.13 H 4.71 N 5.95	79.34 4.75 5.90

In contrast, several methods leading to pyrrolo[3,4—b]pyridines have been reported in the literature. 2,3-Pyridinedicarboxylic acids[7–11], 2,3-pyridinedicarboxylic acid anhydride[12–17], ethyl 2-bromomethylnicotinates[18], 2-hydroxymethylnicotinic acid lactones[19–21], 3-substituted benzoyl-2-pyridinecarboxylic acids[22] and 3-(3-cyano-2-pyridyl)acrylic acids or esters[23] were useful starting materials to prepare substituted 5*H*-pyrrolo[3,4—b]pyridine-5,7 (6*H*)-diones, 6,7-dihydro-5*H*-pyrrolo[3,4—b] pyridine-5-ones, or 6,7-dihydro-5*H*-pyrrolo[3,4—b]pyridines. On the other hand, properly substituted 3-aminopyrroles[24–26] as well as 4-arylmethylene-pyrrolidines[27–29] were employed to construct derivatives of 6,7-dihydro-5*H*-pyrrolo[3,4—b]pyridin-7-ones, 1,6-dihydro-2*H*-pyrrolo[3,4—b]pyridin-2-ones, 6*H*-pyrrolo[3,4—b]pyridines, or 4,5,6,7-tetrahydro-1*H*-pyrido[3,4—b]pyridin-7-ones. Moreover, substituted 4a,7a-dihydro-6-phenyl-1*H*-pyrrolo[3,4—b]pyridine-5,7(4*H*,6*H*)-diones[30] and 3,6-diphenyl-4-morpholino-5*H*-pyrrolo[3,4—b]pyridine-5,7(6*H*)-dione[31] have been prepared by *Diels-Alder* reactions of *N*-phenylmaleimide with substituted cinnamylideneanilines and 6-morpholino-5-phenyl-1,3-oxazin-2-one, respectively.

The possibility of converting pyrano[2,3—c]pyrroles to 5*H*-pyrrolo[3,4—b]pyridines has been realized by preparing 1,6-dimethyl-1,2,3,4,6,7-hexahydro-5*H*-pyrrolo[3,4—b]pyridin-5-one from 6-methyl-3,4,6,7-tetrahydropyrano[2,3—c]pyrrol-5(2*H*)-one and methylamine[5].

Whereas no pharmacological data have been recorded in the literature for pyrano[2,3—c]pyrroles, some substituted pyrrolo[3,4—b]pyridines exhibited hypotensive[12,19,20], tranquilizing[16,32], hypnotic[32], muscle relaxing[32], anticonvulsant[16,17], coronary vasodilatating[27,28], antiinflammatory[33] and prostaglandin synthase[26] inhibitory potencies. Our new compounds may, analogously, show interesting pharmacological activities.

Spectroscopic data of compounds **5** and **6** are recorded in Table 2. In the ^{1}H-NMR (δ ppm) spectra, all the compounds revealed the C-7 proton singlet around 6.4 and the 3-unsubstituted derivatives **6a** and **6b** showed the C-3 proton singlet around 5.6. Whereas, the 4-hydroxyl group signal is not apparent in the spectra of **5**, it is identified in the spectra of **6** as singlet in the 9.0–11 region. The mass spectrum of **5a** is characterized by an intense molecular ion which constitutes the base peak of the spectrum and ions at *m/e* 381, 290 and 278 corresponding to losses of CO, C_6H_5NCO and $C_6H_5CH_2—C=C=O$ from the molecular ion, respectively. The moderately intense ion at *m/e* 181 may be formulated as the radical ion $C_6H_5—N=CH—C_6H_5$. Compound **6a** showed similar fragmentations; however, the base peak of the spectrum is the ion *m/e* 196 and the loss of CO from the parent ion is not observed. In addition, the spectrum revealed an M-C_4H_9 ion at *m/e* 331.

Experimental

Melting points are uncorrected. IR spectra: Perkin-Elmer 421. ^{1}H-NMR spectra: Varian EM 360 with *TMS* as internal standard. Mass spectra: Organic MS 20 AEI (70 eV).

Table 2. *Spectroscopic data of substituted 4-hydroxy-2,5,6,7-tetrahydropyrano[2,3—c]pyrrole-2,5-diones* (**5**) *and 4-hydroxy-1,2,6,7-tetrahydro-5H-pyrrolo[3,4—b]pyridine-2,5-diones* (**6**)

Compound	IR (KBr) cm^{-1}	^{1}H-NMR (*DMSO-d_6*)
5a	3350 m (OH), 1720 m (2-CO), 1685 s, (5-CO), 1610 m, 1590 m, 1485 m, 1455 m, 1420 w, 1370 s.	δ = 3.65 (s, CH_2); 6.28 (s, H at C-7); 6.85–7.50 (m, 15 aromat. H).
5b	3380 m (OH), 1730 m (2-CO), 1690 s (5-CO), 1600 m, 1585 sh, 1495 m, 1455 m, 1375 s.	δ = 2.20 (s, CH_3); 3.65 (s, CH_2); 6.31 (s, H at C-7); 7.00–7.35 (m, 14 aromat. H).
6a	3150–2100 m peaked at 2965 (OH and CH), 1690 s (2-CO), 1650 sh (5-CO), 1620 m, 1545 s, 1490 s, 1450 m, 1410 m, 1365 s.	δ = 0.35–1.40 [m, $CH_3(CH_2)_2$]; 2,18 (s, CH_3); 3.15–3.75 (m, NCH_2); 5.61 (s, H at C-3); 6.5 (s, H at C-7); 6.55–7.35 (m, 9 aromat. H); 10.8 (s, OH).
6b	3105–2200 w (OH), 1700 s (2-CO), 1660 sh (5-CO), 1610 w, 1540 s, 1490 m, 1455 w, 1410 w, 1365 s.	δ = 4.80 (d, NCH_2); 5.62 (s, H at C-3); 6.30 (s, H at C-7); 6.40–7.40 (m, 15 aromat. H); 11.0 (s, OH).
6c	3310 m (OH), 3100–2840 m (CH), 1680 s (2-CO), 1655 m (5-CO), 1630 w, 1595 s, 1585 s, 1495 s, 1455 m, 1410 m, 1395 m.	δ = 1.00 (t, J = 7 Hz, CH_3); 2.20 (s, CH_3); 2.50 (q, CH_2); 4.91 (s, NCH_2); 6.55 (s, H at C-7); 6.60–7.50 (m, 14 aromat. H); 9.72 (s, OH)

6d	3280 m (OH), 3100-2800 m (CH), 1665 s (2-CO), 1650 sh (5-CO), 1625 m, 1605 s, 1585 sh, 1485 s, 1450 m, 1420 s, 1400 s.	δ = 1.22 (d, J = 7 Hz, two CH_3); 2.20 (s, CH_3); 3.31 (q, CH); 4.91 (s, NCH_2); 6.56 (s, H at C-7); 6.65-7.45 (m, 14 aromat. H); 9.50 (s, OH).
6e	3400 m (OH), 3150-2850 m (CH), 1690 s (2-CO), 1670 s (5-CO), 1645-1590 s, 1510 s, 1470 w, 1465 m, 1390 s.	δ = 0.65-1.65 (m, C_4H_9); 6.32 (s, H at C-7); 6.50-7.75 (m, 15 aromat. H); 9.7 (s, OH).
6f	3400 m (OH), 1690 s (2-CO), 1675 s (5-CO), 1640 w, 1595 m, 1500 m, 1475 w, 1460 m, 1390 s.	δ = 3.82 (s, CH_2); 6.25 (s, H at C-7); 6.45-7.70 (m, 20 aromat. H).
6g	3400 (OH), 3100 w, 2960 w, 1690 s (2-CO), 1670 s (5-CO), 1635 m, 1620 sh, 1590 m, 1510 m, 1475 m, 1405 sh, 1380 s.	δ = 6.32 (s, H at C-7); 6.50-7.75 (m, 20 aromat. H); 9.85 (s, OH).

MS of **5a**: *m/e* (relative abundance %) 411 (6, $M+2$), 410 (36, $M+1$), 409 (100, M^+) 381 (11), 317 (6), 316 (27), 290 (6), 289 (18), 279 (5), 278 (20), 277 (6), 276 (5), 252 (10), 183 (29), 182 (66), 181 (39), 180 (40), 131 (8, C_6H_5—C=C=O), 104 (10), 91 (11, $C_6H_5CH_2$), 77 (23, C_6H_5).

MS of **6a**: *m/e* 389 (13, $M+1$), 388 (45, M^+), 371 (10), 333 (11), 332 (38), 331 (27), 275 (8), 256 (15), 255 (40, M-3 —$CH_3C_6H_4NCO$), 219 (5), 197 (20), 196 (100), 195 (13, $CH_3C_6H_4N$=CHC_6H_5), 194 (27), 190 (7), 165 (14), 118 (15), 109 (6), 91 (41, C_7H_7).

1,5-Diphenylpyrrolidine-2,4-diones (**1**)

1,5-Diphenylpyrrolidine-2,4-dione (**1a**) and 1-(3-methylphenyl)-5-phenylpyrrolidine-2,4-dione (**1b**) were prepared as described earlier[1].

1,5-Diaryl-1,5-dihydro-4-substituted amino-2H-pyrrol-2-ones (**3**)

1,5-Dihydro-1,5-diphenyl-4-phenylamino-2*H*-pyrrol-2-one (**3d**) was prepared as previously reported[1]. Analogously, the following compounds were obtained from **1a** or **1b** and the desired amines **2**.

4-Butylamino-1,5-dihydro-1-(3-methylphenyl)-5-phenyl-2H-pyrrol-2-one (**3a**)

Recrystallized from ethanol, m.p. 152 °C; yield 52%.

IR (KBr): 3260 m, 3060 w, 2960 w, 2930 w (NH and CH), 1655 m (C=O), 1600 s, 1540 m, 1490 m, 1450 w shouldered at 1465, 1350 cm^{-1} s.

^{1}H-NMR (*DMSO-d_6*): $\delta = 0.5$-1.6 [m, $CH_3(CH_2)_2$]; 2.12 (s, CH_3); 2.65-3.1 (m, N—CH_2); 4.6 (s, H at C-5); 5.58 (s, H at C-3); 6.2-7.35 (m, NH + 9 aromatic H).

$C_{21}H_{24}N_2O$ (320.4). Calcd. C 78.71, H 7.55, N 8.74.
Found. C 78.90, H 7.70, N 8.40.

4-Benzylamino-1,5-dihydro-1,5-diphenyl-2H-pyrrol-2-one (**3b**)

Recrystallized from ethanol, m.p. 188-189 °C; yield 68%.

IR (KBr): 3230 m, 3050 m (NH), 1650 m (C=O), 1610 s, 1545 m, 1495 m, 1450 w, a band splitted at 1360 m and at 1315 cm^{-1} m.

^{1}H-NMR (*DMSO-d_6*): $\delta = 4.08$ (d, $J = 6$ Hz, CH_2); 4.5 (s, H at C-5); 5.66 (s, H at C-3); 6.5-7.45 (m, NH + 15 aromatic H).

$C_{23}H_{20}N_2O$ (340.4). Calcd. C 81.15, H 5.92, N 8.23.
Found. C 80.92, H 5.71, N 8.27.

4-Benzylamino-1,5-dihydro-1-(3-methylphenyl)-5-phenyl-2H-pyrrol-2-one (**3c**)

Recrystallized from benzene, m.p. 172-173 °C; yield 60%.

IR (KBr): 3300 m, 3100-2800 m with multiple splits (NH and CH), 1665 m (C=O), 1620 s, 1540 m, 1490 m, 1450 w, 1440 w, 1360 cm^{-1} s.

^{1}H-NMR (*DMSO-d_6*): $\delta = 2.15$ (s, CH_3); 4.2 (d, $J = 6$ Hz, CH_2); 4.6 (s, H at C-5); 5.75 (s, H at C-3); 6.5-7.55 (m, NH + 14 aromatic H).

$C_{24}H_{22}N_2O$ (354.5). Calcd. C 81.33, H 6.26, N 7.9.
Found. C 81.65, H 6.36, N 7.7.

3-Benzyl-6,7-diphenyl-4-hydroxy-2,5,6,7-tetrahydro-pyrano[2,3—c]pyrrole-2,5-dione (**5a**)

It was prepared by fusing a mixture of **1a** (0.25 g, 1 mmol) and bis-2,4,6-trichlorphenyl benzylmalonate (**4e**)[34] (0.55 g, 1 mmol) at 160 °C for 1 h in an oil bath. The cooled reddish brown reaction mixture was treated with benzene—petroleum ether 40-60 °C mixture and the separated product was filtered and recrystallized from ethanol, m.p. 246-248 °C; yield 0.17 g (41.5%).

$C_{26}H_{19}NO_4$ (409.4). Calcd. C 76.27, H 4.68, N 3.42.
Found. C 76.0, H 4.89, N 3.60.

3-Benzyl-4-hydroxy-6-(3-methylphenyl)-7-phenyl-2,5,6,7-tetrahydropyrano[2,3—c]pyrrole-2,5-dione (**5b**)

A solution of **1b** (1.3 g, 4.9 mmol) and **4e** (2.71 g, 4.9 mmol) in chlorobenzene (10 ml) was refluxed for 2 h. Subsequently, the cooled reaction mixture was treated with an equal volume of petroleum ether 40-60 °C and the precipitated product was filtered, washed with petroleum ether and recrystallized from ethanol, m.p. 225-228 °C; yield 0.38 g (18.3%).

$C_{27}H_{21}NO_4$ (423.5). Calcd. C 76.58, H 5.0, N 3.31.
Found. C 76.80, H 5.1, N 3.30.

Substituted 4-hydroxy-7-phenyl-1,2,6,7-tetrahydro-5H-pyrrolo[3,4—b]pyridine-2,5-diones (**6**)

An intimate mixture of the appropriate **3** and **4**[34] (1 mmol of each) was heated at 160 °C for 1 h in an oil bath. After cooling, the dark brown reaction mixture was treated with benzene and the resulting solid was filtered, washed with benzene and recrystallized from the suitable solvent. In this manner the following compounds were prepared (for characteristic data refer to Table 1):

1-Butyl-4-hydroxy-6-(3-methylphenyl)-7-phenyl-1,2,6,7-tetrahydro-5H-pyrrolo[3,4—b]pyridine-2,5-dione (**6a**);

1-Benzyl-6,7-diphenyl-4-hydroxy-1,2,6,7-tetrahydro-5H-pyrrolo[3,4—b]pyridine-2,5-dione (**6b**);

1-Benzyl-3-ethyl-4-hydroxy-6-(3-methylphenyl)-7-phenyl-1,2,6,7-tetrahydro-5H-pyrrolo[3,4—b]pyridine-2,5-dione (**6c**)

1-Benzyl-4-hydroxy-3-isopropyl-6-(3-methylphenyl)-7-phenyl-1,2,6,7-tetrahydro-5H-pyrrolo[3,4—b]pyridine-2,5-dione (**6d**)

3-Butyl-4-hydroxy-1,2,6,7-tetrahydro-1,6,7-triphenyl-5H-pyrrolo[3,4—b]pyridine-2,5-dione (**6e**)

3-Benzyl-4-hydroxy-1,2,6,7-tetrahydro-1,6,7-triphenyl-5H-pyrrolo[3,4—b]pyridine-2,5-dione (**6f**)

4-Hydroxy-1,2,6,7-tetrahydro-1,3,6,7-tetraphenyl-5H-pyrrolo[3,4—b]pyridine-2,5-dione (**6g**)

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