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Gould-Jacobs Reaction of 6-Amino-2,3-diphenylquinoxaline#

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Summary. Catalytic hydrogenation of 2,3-diphenyl-6-nitroquinoxaline led to the corresponding amine **1** which in turn afforded products **3a–i** on reaction with alkoxymethylene derivatives **2a–i**. Thermal cyclization of **3b** and **3f** yielded substituted pyrazinoquinolones **5b** and **5f**, respectively. Optimal conditions for the successful hydrolysis of ester **5b** were found. The structures of all products were deduced from their IR, UV, ^{1}H, and ^{13}C NMR spectroscopic data.

Keywords. Pyrazinoquinolones; Quinoxalinoaminoethylenes; 6-Aminoquinoxalines; ^{1}H and ^{13}C NMR; Biological activity.

***Gould-Jacobs*-Reaktion von 6-Amino-2,3-diphenylchinoxalin**

Zusammenfassung. Katalytische Hydrierung von 2,3-Diphenyl-6-nitrochinoxalin ergibt das entsprechende Amin **1**, welches bei Reaktion mit den Alkoxymethylenderivaten **2a–i** zu den Produkten **3a–i** führt. Thermische Cyclisierung von **3b** und **3f** liefert die substituierten Pyrazinochinolone **5b** und **5f**. Für die Hydrolyse des Esters **5b** wurden optimale Bedingungen ermittelt. Die Strukturen aller Produkte wurden aus spektroskopischen Daten hergeleitet (IR, UV, ^{1}H- und ^{13}C-NMR).

Introduction

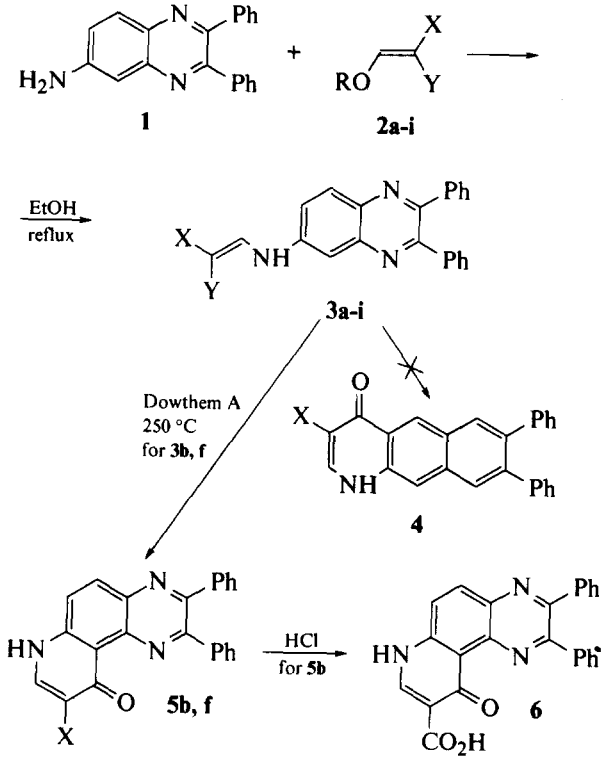
The alkoxy group in alkoxymethylene derivatives of diesters or dinitriles of malonic acid, 2,4-pentanedione, alkyl cyanoacetate, or alkyl acetoacetate can easily be replaced by a suitable nucleophile (e.g. aniline or heterocyclic amines). These substitution products can be cyclized to form a 4-pyridone ring fused to the starting aromatic or heteroaromatic skeleton [1]. The 6-substituted aminoquinoxalines furnish angularly annelated substituted pyrazinoquinolones [2] under the conditions of the *Gold-Jacobs* reaction [3]. This paper presents the preparation of 2,3-diphenylquinoxaline derivatives with an aminoethylene substituent in position 6 and two electron-accepting groups (nitrile, acetyl, alkoxycarbonyl, or their combi-

Dedicated to Prof. *F. Sauter* on the occasion of his 65th birthday

nation); the condition for the thermal cyclization of **3b** and **3f** and the hydrolysis of **5b** have been examined as well.

Results and Discussion

The starting material for the synthesis of 6-substituted derivatives **3** was 2,3-diphenyl-6-nitroquinoxaline, obtained by reaction of benzil with 4-nitro-1,2-phenylenediamine [4]. Catalytic hydrogenation of the nitro group (*Raney* nickel in ethanol) yielded the respective amine (**1**) which was then subjected to the reaction with alkoxymethylene derivatives **2** (Scheme 1) obtained by the condensation of alkyl orthoformate with an active methylene component (derivatives of malonic and 3-oxobutanoic acid, 2,4-pentadione) [5–7].



1 **2a-i** **3a-i** **4** **5b, f** **6**

	X	*Y*	*R*
a	CO_2CH_3	CO_2CH_3	CH_3
b	$CO_2C_2H_5$	$CO_2C_2H_5$	C_2H_5
c	CN	CN	C_2H_5
d	$COCH_3$	$COCH_3$	C_2H_5
e	$COCH_3$	CO_2CH_3	CH_3
f	$COCH_3$	$CO_2C_2H_5$	C_2H_5
g	$COCH_3$	CN	C_2H_5
h	CN	CO_2CH_3	CH_3
i	CN	$CO_2C_2H_5$	C_2H_5

Scheme 1

Thermal cyclocondensation of **3b** and **3f** in an inert solvent at 260 °C afforded angularly fused 5,8-dihydro-2,3-diphenyl-7-ethoxycarbonyl-8-oxopyrido[3,2-*f*]-quinoxaline (**5b**) and 5,8-dihydro-2,3-diphenyl-7-acetyl-8-oxopyrido[3,2-*f*]quinoxaline (**5f**), respectively. Formation of linearly annelated products **4** was not observed (Scheme 1).

Acid hydrolysis of **5b** [8] (concentrated hydrochloric acid) yields acid **6** (Scheme 1) which can be considered as an analogue of nalidixic acid nonalkylated at the pyridone nitrogen atom. Basic hydrolysis [9] does not lead to free acid, probably caused by the low solubility of the starting ester in the aqueous medium. The yields and physico-chemical data of all compounds are listed in Table 1.

The UV spectrum of 2,3-diphenylquinoxaline presents three absorption bands [10] (λ_{max}, $\log \varepsilon$): 244 (3.53), 265 (3.36), and 345 (3.07) nm. Introduction of a polar aminoethylene substituent into the 2,3-diphenylquinoxaline skeleton resulted in a shift of the most intense maximum to 312 nm (Table 2). The longest wavelength band is bathochromically shifted by 40 nm, indicating strong conjugation between the substituent and the ring. The same phenomenon was observed for the cyclized derivatives **5b**, **5f**, and **6** (Table 2).

The IR spectra of compounds **3** revealed stretching vibrations of the cyano group at 2220 or 2210 cm^{-1} and CH and NH bonds at 2950–3160 and 3445 cm^{-1}, respectively (Table 3).

Derivatives **3e–i** can exist as two isomers with respect to their asymmetric substitution (*X*, *Y*) at the aminoethylene residue. The relative ratio of the individual geometric isomers could be estimated from their NMR spectral data considering the integral intensities of suitable signals. For instance, the *E*:*Z* ratio for **3g** was found to be 1:1, whereas with other compounds the energetically more favoured *E* isomers prevailed. The *E*:*Z* ratio of derivatives with a cyano group (**3h, i**) amounted to 60:40, that of derivatives with the bulkier acetyl group (**3e, f**) to 90:10. This can

Table 1. Physico-chemical data of compounds **3a–i**, **5b, f** and **6**[a]

	Formula	Mol. Weight	M.p. (°C)	Yield (%)
3a	$C_{26}H_{21}N_3O_4$	439.5	173–175	73
3b	$C_{28}H_{25}N_3O_4$	467.5	164–165	78
3c	$C_{24}H_{15}N_5$	373.4	293–296	86
3d	$C_{26}H_{21}N_3O_2$	407.5	311–313	67
3e	$C_{26}H_{21}N_3O_3$	423.5	223–225	55
3f	$C_{27}H_{23}N_3O_3$	437.5	159–160	61
3g	$C_{25}H_{18}N_4O$	390.4	232–234	53
3h	$C_{25}H_{18}N_4O_2$	406.4	259–261	56
3i	$C_{26}H_{20}N_4O_2$	420.5	248–250	58
5b	$C_{26}H_{19}N_3O_3$	421.5	216–218	58
3f	$C_{25}H_{17}N_3O_2$	391.4	244–247	48
6	$C_{24}H_{15}N_3O_3$	393.4	261–265	81

[a] All elemental analyses (C, H, N) are in good agreement with the calculated values

most probably be explained by the existence of intramolecular hydrogen bonding between the acetyl group and the imino hydrogen. This bond, however, does not exist in *E* isomers of cyano derivatives. For acetoacetate derivatives, this intramolecular bond is stronger with the carbonyl oxygen atom of the acetyl group than with that of the ester group. The presence of hydrogen bonding is demonstrated by by $^3J_{CH-NH}$ values of about 12–14 Hz (Table 4), the chemical shift of carbonyl carbons (at higher field in comparison with "free" carbonyls, Table 6), and ν(C=O) frequencies for ester and acetyl groups at 1680 cm^{-1} and 1630 cm^{-1} (Table 3), respectively.

The strong polarizations of the aminoethylene residue was reflected by the shift of the C-10 signal in the ^{13}C NMR spectrum (Table 6). When the ethylene residue was substituted by two equal substituents, the most electron-accepting groups turned out to be nitriles (**3c**, $\delta = 53.8$ ppm); worse acceptors were akloxycarbonyl

Table 2. UV spectroscopic data of compounds **3a–i**, **5b**, **f**, and **6**

	λ_{max} (log ε) (nm)
3a	264 (3.27), 312 (3.53), 320 (3.51), 386 (3.51)
3b	227 (3.52), 265 (3.27), 312 (3.54), 325 (3.50), 388 (3.51)
3c	231 (3.58), 267 (3.50), 310 (3.71), 324 (3.66), 381 (3.77)
3d	234 (3.40), 263 (3.39), 316 (3.35), 387 (3.56)
3e	233 (3.36), 265 (3.17), 316 (3.22), 387 (3.42)
3f	232 (3.52), 264 (3.31), 316 (3.36), 387 (3.58)
3g	236 (3.56), 267 (3.47), 315 (3.40), 386 (3.74)
3h	234 (3.31), 264 (3.19), 312 (3.44), 326 (3.40), 383 (3.55)
3i	231 (3.48), 265 (3.31), 312 (3.56), 325 (3.52), 384 (3.69)
5b	236 (3.89), 256 (3.98), 293 (3.74), 387 (3.67)
5f	236 (3.61), 260 (3.66), 296 (3.31), 375 (3.23), 388 (3.29)
6	232 (3.79), 251 (3.90), 266 (3.84), 385 (3.58)

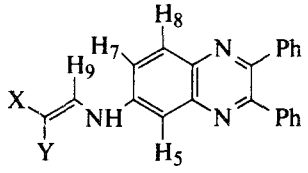
Table 3. IR spectroscopic data of compounds **3a–i**, **5b**, **f**, and **6**

	ν (cm^{-1})
3a	3320, 3070, 1635, 1620
3b	3270, 3070, 2980, 1695, 1655, 1610
3c	3220, 3070, 2220, 2210, 1650, 1600
3d	3050, 1635, 1620, 1580
3e	3060, 2950, 1715, 1640, 1620, 1580
3f	3070, 2980, 1715, 1640, 1620, 1580
3g	3180, 3060, 2210, 1680, 1650, 1620
3h	3050, 2950, 2210, 1720, 1660, 1640, 1610
3i	3190, 3050, 2980, 2210, 1715, 1685, 1635, 1610
5b	3060, 2976, 1728, 1695, 1616
5f	3056, 1676, 1644, 1616
6	3056, 1620, 1609, 1576

(**3a, b**; $\delta = 95.4$ ppm and 95.9 ppm, respectively), the least accepting ones were acetyl groups (**3d**, $\delta = 114.9$ ppm). The most polarized substituents with unequal groups were cyanoacetates (**3h, i**), followed by 3-oxobutanenitrile (**3g**) and 3-oxobutanoic acid esters (**3e, f**).

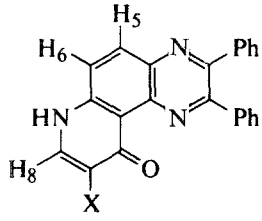
The ^{13}C NMR signals of all compounds (Tables 6, 7) were assigned using the APT technique and by comparison of observed values with calculated ones (obtained by superposition of 2,3-diphenylquinoxaline [11] with either the

Table 4. ^1H NMR spectroscopic data of compounds **3a–i** (δ; ppm; J: Hz)



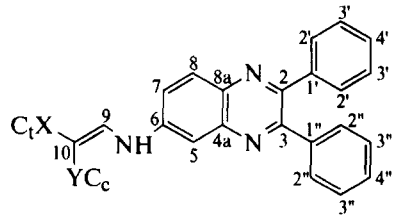
	H_5	H_7	H_8	H_9	NH	H_{Ph}	$J_{9,NH}$	$J_{7,8}$	$J_{7,5}$	other signals
3a	8.13	7.96	8.04	8.62	10.86	7.32–7.46	14.1	8.7	1.8	3.71, 3.78
3b	8.04	7.96	8.13	8.58	10.88	7.32–7.49	13.5	9.0	2.1	4.16, 1.27, 4.25, 1.28
3c	8.20	8.01	8.15	8.84	11.50	7.33–7.48		9.1	2.3	
3d	8.32	8.16	8.03	8.64	12.61	7.32–7.48	12.3	9.0	1.8	2.43, 2.47
3e	7.98	7.60	8.20	8.73	12.96	7.32–7.56	12.9	9.0	2.4	3.85, 2.63
	7.96			8.81	10.61					3.94, 2.53
3f	8.09	7.95	8.14	8.64	12.63	7.31–7.47	12.9	9.0	2.4	2.46, 4.20, 1.31
	8.06			8.58	10.85					2.42, 4.30, 1.33
3g	8.17	8.08	8.23	8.65	11.06	7.33–7.50	13.5	9.3	2.4	2.40
			8.30	8.73	12.25		13.2		2.4	2.37
3h	8.07	7.99	8.14	8.59	11.19	7.32–7.48	13.8	9.3	2.4	3.76
	8.26	8.06		8.78	10.96			8.7	2.4	3.81
3i	8.07	7.99	8.18	8.59	11.19	7.32–7.48	13.9	9.0	2.4	4.23, 1.29
	8.26	8.09		8.76	10.97		13.6		2.4	4.25, 1.30

Table 5. ^1H NMR spectroscopic data of compounds **5b, f** and **6** (δ: ppm; J: Hz)



	H_5	H_6	H_8	H_{Ph}	$J_{5,6}$[Hz]	other signals
5b	8.41	8.51	9.14	7.3–7.7	6.0	4.76, 1.51
5f	8.33	8.39	9.37	7.4–7.7	9.0	2.90
6	8.07	8.37	8.94	7.3–7.7	9.0	

Table 6. ^{13}C NMR spectroscopic data of compounds **3a–i** (δ: ppm)



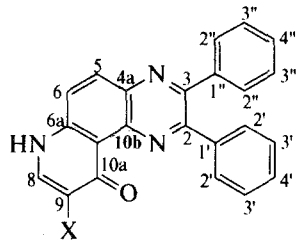
C	3a	3b	3c	3d	3e	3f	3g	3h	3i
2	154.5	154.6	152.4	a	154.8	154.9	154.8	154.1	154.9
3	151.8	152.6	151.7	a	154.8	154.7	154.7	153.6	154.8
4a	131.8	a	132.6	133.9	134.9	134.9	135.5	134.4	133.7
5	138.2	138.9	138.0	138.9	137.7	137.7	137.9	137.9	137.7
6	110.9	112.8	113.6	111.9	a		a	112.6	110.9
7	130.1	131.1	130.2	130.2	130.3	130.3	132.3	130.5	129.8
8	123.7	121.9	131.9	124.7	124.4	124.3	124.4	123.5	124.4
8a	141.8	142.1	140.6	141.4	140.7	140.8	140.2	140.4	140.8
9	149.1	150.8	153.8	154.6	147.1	147.1	147.0	151.6	147,1
10	95.4	95.9	53.8	114.9	a		a	75.8	76.6
1′, 1″	138.9	140.4	138.2	138.8	138.2	138.2	138.0	138.4	138.2
2′, 2″	128.2	128.3	128.1	127.8	128.3		129.6	128.0	128.3
	128.4	128.4	128.0	128.4	128.5		129.5		128.5
3′, 3″	129.8	129.8	129.5	129.8	129.9		129.8	129.5	129.6
	130.8		129.6	130.2	129.7		129.7	129.7	130.3
4′, 4″	129.2	128.9	129.0	128.7	129.8		129.4	128.9	129.0
	129.3	129.1	129.1	129.1	129.6		129.3	129.1	129.6
X, *Y*	51.7	60.8		32.2	58.4		31.7	51.8	58.4
	51.6	60.7		27.5	29.7		28.2		14.0
		14.5							
		14.3							
C_t	167.9	168.9	115.8	199.0	161.4			166.4	166.2
C_c	166.3	165.1	113.3	195.3	194.5	164.6	194.8	117.6	117.8

[a] not observed

aminoethylene substituent [12, 13] or with 3-substituted derivatives of 1,4-dihydro-4-oxopyridine [14]).

The electron impact mass of compound **3** showed intense molecular ion peaks (Table 8). The origin of more intense signals could be explained by bond fission, mainly in the aminoethylene chain. Molecular ions of derivatives with an ester group (**3a, b, e, f, h, i**) loose corresponding alcohol residues and carbon oxide. The formation of the fragment ions was verified by the detection of metastable transitions. The existence of various fragment ions resulting from rupture of the N–C bond in β-position with respect to the ring depends on the localization of the charge on the molecular ion. In the case of derivative **3d**, the base peak is the fragment ion at $m/z = 296$ which is formed by elimination of 3-methylene-2,4-pentandione (β-elimination without rearrangement of hydrogen). Introduction of more electron-

Table 7. ^{13}C NMR spectroscopic data of compounds **5b**, **f** and **6**



C	**5b**	**5f**	**6**
2	151.7	152.6	151.5
3	152.1	153.6	152.1
4a	136.8	137.1	137.6
5	134.9	131.8	134.9
6	121.0	123.2	123.3
6a	142.4	140.6	143.1
8	147.7	154.2	143.7
9	112.3	112.2	112.2
10	178.2	a	178.6
10a	119.6	118.9	118.0
1a	138.3	140.0	139.1
1′, 1″	138.2, 139.1	137.9	138.4
2′, 2″	128.0, 128.2	128.6, 128.9	128.2, 128.3
3′, 3″	129.5	128.7	129.7
4′, 4″	129.0	129.5	129.1
C=O	166.0	197.5	166.5
others	50.1, 14.8	32.3	

[a] not observed

Table 8. Mass spectroscopic data of compounds **3a**, **b**, **d**, **f–i**, **5f**, and **6**

	m/z (%)
3a	440 $(M + 1)^+$ (24), 439 M^+ (85), 408 (31), 407 (100), 348 (29), 319 (15), 142 (19), 115 (15), 63 (22)
3b	467 M^+ (12), 421 (13), 298 (17), 297 (83), 296 (73), 91 (49), 64 (16), 63 (10), 46 (46), 43 (100)
3d	408 $(M + 1)^+$ (27), 407 M^+ (99), 390 (19), 364 (35), 322 (19), 297 (36), 296 (100), 112 (35), 77 (22), 43 (50)
3f	438 $(M + 1)^+$ (30), 437 M^+ (100), 391 (22), 364 (19), 363 (62), 349 (21), 348 (78), 301 (21), 296 (26), 285 (18), 43 (16)
3g	391 $(M + 1)^+$ (28), 390 M^+ (100), 389 (30), 348 (11), 347 (29), 282 (11), 141 (13), 114 (11), 63 (12), 43 (25)
3h	407 $(M + 1)^+$ (29), 406 M^+ (100), 375 (12), 374 (42), 373 (61), 346 (11), 167 (10), 139 (14), 117 (10), 63 (13)
3i	421 $(M + 1)^+$ (31), 420 M^+ (100), 375 (15), 374 (47), 373 (60), 348 (8), 347 (19), 345 (9), 140 (12), 63 (16)
5f	391 M^+ (46), 376 (100), 270 (14), 186 (10), 77 (14), 43 (8)
6	350 (12), 349 (42), 348 (33), 115 (16), 44 (100)

drawing substituents causes higher polarization of the double bond, indicated by decreasing intensity of the ion at $m/z = 296$ and 297 (β-elimination either with or without rearrangement of hydrogen [7]). In the mass spectra of acid **6**, the peak of the molecular ion could not be observed. The base peak is the fragment ion at $m/z = 44$ (carbon dioxide) which is formed by decarboxylation (Table 8).

Biological activity

We were also interested in the potential antimicrobial activity of the synthesized compounds. The basic antimicrobial screening was realized on the standard set of gram-negative bacterial strains (*Bacillus subtilis* CCM 2216, *Escherichia coli* CNCTC 326/71, *Serratia marcescens* CCM 303, *Pseudomonas aeruginosa* CNCTC 133/71, *Staphylococcus aureus* CNCTC 78/71, *Entherococcus faecalis* CCM 1875) and one yeast microorganism (*Saccharomyces cerevisiae* DT XII). The bacterial strains were taken from the collections of microorganisms CNCTC (Czechoslovak National Collection of Type Cultures, Prague) and CCM (Czechoslovak Collection of Microorganisms, Brno). *S. cerevisiae* was purchased from the Department of Microbiology and Virology, Faculty of Natural Sciences, Comenius University, Bratislava.

Antimicrobial activity was characterized by the MIC values (mininal inhibitory concentration). The MIC was determined by a macrodilution method, using *Mueller–Hinton* broth for the bacteria and *Sabouraud* broth for *S. cerevisiae*. The final concentration of inoculum was approximately 5×10^5 cfu/ml.

For the experiments, the substances were dissolved in *DMSO* (500 μg/ml); further dilutions in the test medium, using a twofold dilution series, gave the required concentrations (generally in the 250 – 1 μg/ml range). The MIC (Table 9) was defined as the lowest concentration of compound that completely inhibited growth after 24 hours incubation at 37 °C for the bacteria or 28 °C for *S. cerevisiae*.

Table 9. MIC for compounds **3a–i**, **5b**, **f**, and **6**

	1	2	3	4	5	6	7
3a	>250	>250	>250	>250	>250	>250	>125
3b	>250	>250	>250	>250	>250	>250	>125
3c	>250	>250	>250	>250	>250	>250	>250
3d	>250	>250	>250	>250	>250	>250	>250
3e	>250	>250	>250	>250	>250	>250	>250
3f	>250	>250	>250	>250	>250	>250	>250
3g	>250	>250	>250	>250	>250	>125	>125
3h	>250	>250	>250	>250	>250	>250	>250
3i	>250	>250	>250	>250	>250	>250	>250
5b	>250	>250	>250	>250	>250	>250	>250
5f	>250	>250	>250	>250	>250	>250	>250
6	>250	>250	>250	>250	>250	>250	>250

1: *Escherichia coli* 326/71; 2: *Serratia macrescens* 303; 3: *Pseudomonas aeruginosa* 133/71, 4: *Bacillus subtilis* 2216, 5: *Enterococcus faecalis* 1875, 6: *Staphylococcus aureus* 78/71, 7: *Saccharomyces cerevisiae* DT XII

Experimental

The melting points were measured on a Kofler micro hot-stage. The IR spectra (0.5 mg of the substance per 300 mg KBr) and the UV spectra (saturated solutions in methanol, cell width 2 mm) were recorded with FTIR PU 9802 (Philips) and Specord (Zeiss, Jena) spectrophotometers, respectively. The ^1H and ^{13}C NMR spectra of *DMSO*-d_6 solutions were measured with a Varian VXR-300 instrument at 298 K in a 5 mm multinuclear probe δ in ppm relative to internal *TMS*). The ^1H NMR spectra were recorded at a spectral width of 4 kHz; number of data points: 16 k. The ^{13}C NMR spectra were measured at 75 MHz; spectral width: 16 kHz, 64 k data points. The number of accumulations for proton decoupled ^{13}C NMR spectra varied within 250 and 5000. The pulse repetition time was 3 s, the flip angle 45°. The electron impact mass spectra were taken with an MS 902S (AEI-Kratos) instrument at 70 eV electron energy and 100 μA trap current.

6-Substituted aminoethylene derivatives of 2,3-diphenylquinoxaline (**3a–i**)

2,3-Diphenyl-6-nitroquinoxaline (10 mmol) in ethanol was hydrogenated at 120 kPa on *Raney* nickel until 660 ml of hydrogen had been consumed. The catalyst was filtered off, the respective alkoxymethylene derivative (**2a–i**, 10 mmol) was added, and the mixture was refluxed for 30 min. The mixture was shortly boiled with charcoal, filtered, the major part of solvent was evaporated, and the separated product was filtered off and washed with cold ethanol. Crystallization from ethanol afforded analytically pure products.

5,8-Dihydro-2,3-diphenyl-7-ethoxycarbonyl-8-oxopyrido[3,2-f]quinoxaline (**5b**)

Compound **3b** (1 g) and Dowtherm (15 ml) were heated at 260 °C for 4 h. The precipitate formed after cooling was collected by suction and washed with diethyl ether. The cyclizate was recrystallized from *DMF* with addition of charcoal. The collected and washed precipitate was dried in vacuum at 80 °C for 4 h.

5,8-Dihydro-2,3-diphenyl-7-acetyl-8-oxopyridol[3,2-f]quinoxaline (**5f**)

Compound **3f** (1 g) and Dowtherm (50 ml) were heated at 260 °C for 6 h. The mixture was cooled, hexane (100 ml) was added, the separated precipitate was filtered off, washed with diethyl ether, and recrystallized from *DMF*/water with addition of charcoal. The collected and washed precipitate was dried in vacuum at 80 °C for 4 h.

5,8-dihydro-2,3-diphenyl-8-oxopyrido[3,2-f]quinoxaline-7-carboxylic acid (**6**)

A mixture of ethyl ester **5b** (1 g) and concentrated hydrochloric acid (20 ml) were refluxed for 30 min. During the reaction, the major part of the ethyl ester dissolved and, after a certain time, the acid formed began to separate. The reaction mixture was cooled and the precipitate was collected by suction and washed with 20% sodium carbonate solution. The acid **6** was recrystallized from *DMF* with addition of charcoal.

Yields and physico-chemical properties of all compounds are summarized in Table 1.

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