GOULD–JACOBS REACTION OF 5- AND 6-AMINO-2-SUBSTITUTED BENZOXAZOLES. II. REACTION WITH 3-ETHOXYMETHYLENE-2,4-PENTANEDIONE AND ETHYL 2-ETHOXYMETHYLENE-3-OXOBUTANOATE

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Nucleophilic reaction of 5-amino- and 6-amino-2-substituted benzoxazoles **1** and **2** with 3-ethoxymethylene-2,4-pentanedione (**3**) gave compounds **5** and **6**. Compounds **1** and **2** reacted with ethyl ethoxymethylene-3-oxobutanoate (**4**) under formation of compounds **7** and **8**. The substitution products **7** and **8** underwent thermal cyclization at high temperature (boiling mixture of diphenyl ether and biphenyl) to give angularly and linearly annelated derivatives of 5-acetyl-4-oxo-oxazolo[4,5-*f*]-quinoline **9** and 7-acetyl-8-oxo-oxazolo[5,4-*g*]quinoline **10** (from **7**), and derivatives of 8-acetyl-9-oxo-oxazolo[5,4-*f*]quinoline **11** and 6-acetyl-5-oxo-oxazolo[4,5-*g*]quinoline **12** (from **8**). The structure of the substitution products is discussed on the basis of their spectral characteristics (^{1}H and ^{13}C NMR, IR, UV, MS).

Key words: Gould–Jacobs reaction; Oxazoloquinolones.

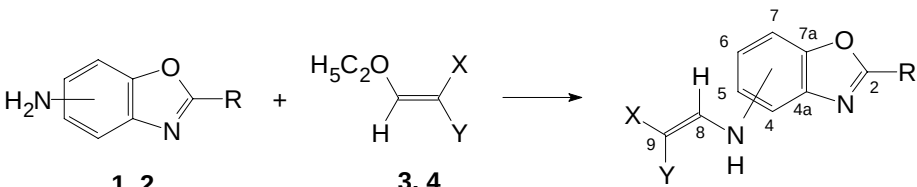
The Gould–Jacobs reaction[1], in which a 4-pyridone ring is annelated to a benzoxazole skeleton[2], represents one of the synthetical approaches to the oxazoloquinolone system. Much more common is the formation of oxazole skeleton on a substituted quinoline derivative[3–5]. With isoelectric azoles, the former method leads exclusively to angularly annelated derivatives, with nonisoelectric ones it enables the preparation of both the linearly and angularly annelated products, the choice of the substitution derivatives being, however, more difficult.

The present study represents an extension of our preceding paper[6] describing the reaction of 5- and 6-amino-2-substituted benzoxazoles (with hydrogen, methyl or phenyl in position 2) with diethyl ethoxymethylenemalonate leading to 3-*N*-[5- or 6-(2-substituted benzoxazolyl)]aminomethylenemalonates which under conditions of Gould–Jacobs reaction were cyclized to give a mixture of angularly and linearly annelated oxazoloquinolones.

The present communication describes the nucleophilic substitution reaction of 5-amino- (**1**) and 6-amino-2-substituted (**2**) benzoxazoles with 3-ethoxymethylene-2,4-pentanedione (**3**), leading to products **5** and **6**, and with ethyl 2-ethoxymethylene-3-oxobutanoate (**4**), affording products **7** and **8** (Scheme 1). This nucleophilic substitution proceeds under mild conditions, in refluxing ethanol. Generally, the 6-amino-substituted benzoxazoles **2** required longer reaction time, the substituent in position 2 having no marked influence on the reaction course.

The physicochemical properties of the products **5**–**8** are given in Table I and their IR and UV spectra in Table II. In all cases, the substitution derivatives showed transoid arrangement of the –NH–CH= grouping (J = 12–14 Hz). Comparison of ^{1}H NMR signals of compounds **5**–**8** is given in Table III. The position of the NH proton signal was approximately constant (12.57–12.75 ppm) and the signal of the neighbouring C–H group was at 8.30–8.31 ppm. The acetyl signal appeared at 2.10–2.40 ppm and those of the ethyl ester group at 1.24–1.29 and 4.10–4.13 ppm.

The ^{13}C NMR spectra of products **5**–**8** (Table IV) show that in the case of unsymmetrically substituted ethylenic bond both geometric isomers are formed, the (*E*)-isomer invariably predominating (more than 90%). The shift of the acetyl carbons C-10 and C-12 is enormously high, being 199.50–199.38 and 195.63–195.20 ppm, respectively, in compounds **5** and **6**. In the spectra of derivatives **7** and **8** the signal of the C-12 carbon atom, bearing the ester group, appears mostly at 166.00–166.17 ppm. The carbon atom C-2 in derivatives **5a**–**8a** resonates at 151.13–155.49 ppm, in the other derivatives at 162.60–166.22 ppm.



1, 2 **3, 4** **5-8**

In formulae **1**, **5**, **7** : 5-NH_2

2, **6**, **8** : 6-NH_2

3, **5**, **6** : X = Y = $COCH_3$

4, **7**, **8** : X = $COCH_3$, Y = $COOC_2H_5$

1, **2**, **5-8** : **a**, R = H

b, R = CH_3

c, R = C_6H_5

SCHEME 1

TABLE I
Physicochemical properties of substitution products **5**–**8**

Compound	Solvent of crystallization Reaction time, min	M.p., °C Yield, %	Formula M.w.	Calculated/Found		
				% C	% H	% N
5a	toluene	142–143	$C_{13}H_{12}N_2O_3$	63.91	4.95	11.47
	15	70	244.1	63.55	4.78	11.25
5b	toluene	132–135	$C_{14}H_{14}N_2O_3$	65.09	5.47	10.85
	15	73	258.1	65.07	5.48	10.76
5c	toluene	170–171	$C_{19}H_{16}N_2O_3$	71.22	5.04	8.75
	30	75	320.1	70.93	5.03	8.74
6a	toluene	170–171	$C_{13}H_{12}N_2O_3$	61.29	5.15	10.22
	30	67	244.1	61.09	5.31	10.28
6b	toluene	132–134	$C_{14}H_{14}N_2O_3$	62.48	5.60	9.72
	30	65	258.1	62.33	5.54	9.81
6c	toluene	197–199	$C_{19}H_{16}N_2O_3$	68.55	5.18	8.00
	60	65	320.1	68.72	5.19	7.97
7a	$CHCl_3$–hexane	114–117	$C_{14}H_{14}N_2O_4$	61.29	5.15	10.22
	15	80	274.1	61.30	5.09	10.19
7b	toluene	103–106	$C_{15}H_{16}N_2O_4$	62.48	5.60	9.72
	30	78	288.1	62.35	5.69	9.80
7c	toluene	134–135	$C_{20}H_{18}N_2O_4$	68.55	5.18	8.00
	60	82	350.1	68.31	5.15	8.08
8a	$CHCl_3$–hexane	70–71	$C_{14}H_{14}N_2O_4$	61.29	5.15	10.22
	30	65	274.1	61.18	5.20	10.29
8b	toluene	208–211	$C_{15}H_{16}N_2O_4$	62.48	5.60	9.72
	60	60	288.1	62.40	5.54	9.66
8c	toluene	173–175	$C_{20}H_{18}N_2O_4$	68.55	5.18	8.00
	60	62	350.1	68.60	5.12	8.12

TABLE II
Infrared and UV spectra of substitution products **5–8**

Compound	IR spectra		UV spectra
	$\tilde{\nu}$(C=O)[a]	$\tilde{\nu}$(C=C)	
5a	1 618	1 585	206.6 (3.24), 239.9 (3.12), 335.1 (3.34)
5b	1 630	1 574	207.0 (3.72), 241.3 (3.23), 336.0 (3.41)
5c	1 618	1 591	206.3 (3.70), 266.5 (3.42), 341.5 (3.53)
6a	1 637	1 591	203.9 (3.22), 245.1 (3.02), 341.5 (3.40)
6b	1 630	1 574	207.0 (3.72), 241.3 (3.21), 336.9 (3.39)
6c	1 614	1 583	204.9 (3.74), 337.1 (3.60)
7a	1 684, 1 610	1 583	206.3 (3.21), 235.8 (3.33), 334.2 (3.36)
7b	1 711, 1 639	1 581	207.0 (3.22), 236.7 (3.36), 335.1 (3.36)
7c	1 691, 1 633	1 583	201.6 (3.50), 241.3 (3.31), 270.6 (3.33), 339.7 (3.56)
8a	1 695, 1 618	1 581	202.9 (3.17), 232.3 (3.11), 341.5 (3.26)
8b	1 691, 1 631	1 574	204.9 (3.32), 241.3 (3.18), 348.2 (3.40)
8c	1 709, 1 637	1 581	200.0 (3.56), 232.8 (3.30), 379.0 (3.66)

[a] The long-wavenumber absorption corresponds to the ester carbonyl vibration.

TABLE III
Proton NMR spectra of substitution products **5–8**

Compound	H-4	H-5	H-6	H-7	H-8	NH	$COCH_3$	$COCH_3$	CH_2	CH_3	R
5a	7.74	–	7.52	7.70	8.44	12.64	2.38	2.38	–	–	8.76
5b	7.70	–	7.38	7.62	8.40	12.75	2.41	2.37	–	–	2.61
5c	7.79	–	7.50	7.79	8.45	12.62	2.40	2.41	–	–	–[a]
6a	7.80	7.50	–	8.10	8.46	12.64	2.43	2.40	–	–	8.73
6b	7.65	7.42	–	7.79	8.42	12.63	2.39	2.38	–	–	2.60
6c	7.79	7.49	–	8.09	8.46	12.64	2.10	2.10	–	–	–[b]
7a	7.86	–	7.46	7.76	8.46	12.61	2.37	–	4.10	1.29	8.72
7b	7.72	–	7.38	7.66	8.45	12.60	2.37	–	4.18	1.28	2.60
7c	7.90	–	7.46	7.82	8.49	12.62	2.38	–	4.17	1.29	–[c]
8a	7.75	7.34	–	7.85	8.35	12.61	2.38	–	4.15	1.27	8.66
8b	7.85	6.97	–	7.89	8.30	12.57	2.12	–	4.14	1.24	2.60
8c	7.82	7.73	–	7.98	8.51	10.90	2.30	–	4.10	1.24	–[d]

Chemical shifts of *o*- and (*m*-, *p*-)protons, respectively: [a] 8.19, 7.62. [b] 8.15, 7.61. [c] 8.19, 7.62. [d] 8.15, 7.60.

TABLE IV
^{13}C NMR spectra of substitution products **5**–**8**

Compound	C-2	C-3a	C-4	C-5	C-6	C-7	C-7a	C-8	C-9	C-10	C-11	C-12	C-13	C-14	R
5a	155.49	140.70	111.75	136.56	109.45	117.48	147.03	153.53	112.52	195.28	27.48	199.38	31.49	–	–
5b	164.96	141.98	110.52	135.88	108.36	115.67	147.83	152.70	112.50	194.63	28.99	198.94	30.72	–	13.68
5c	163.56	142.50	111.46	136.72	109.09	117.11	147.92	153.24	112.70	195.20	27.40	199.30	31.30	–	–[a]
6a	154.54	137.03	120.60	116.40	137.45	101.10	150.11	153.03	112.81	195.34	27.51	199.50	31.50	–	–
6b	165.40	136.11	116.11	110.91	142.13	108.60	148.05	153.50	112.40	195.20	27.50	199.30	31.50	–	14.17
6c	162.60	137.23	120.24	116.53	138.86	100.77	150.92	152.70	112.80	195.30	27.50	199.50	31.50	–	–[b]
7a	155.20	140.50	109.30	136.30	109.00	111.00	146.90	152.32	102.17	197.92	29.83	166.00	59.40	13.98	–
7b	165.35	142.00	108.57	135.95	110.90	115.70	148.07	152.44	101.91	198.02	30.14	166.16	59.60	14.03	24.15
7c	163.51	142.40	109.02	136.53	111.60	116.70	147.70	152.35	102.07	198.06	30.46	166.14	59.31	14.16	–[c]
8a	151.83	137.15	115.70	129.86	100.89	120.67	150.05	154.26	102.42	198.31	30.50	166.08	59.42	14.15	–
8b	166.22	130.48	114.04	127.45	111.50	116.06	145.46	151.77	101.04	197.93	30.55	169.16	59.27	14.28	23.70
8c	164.21	138.74	115.98	136.86	111.98	120.11	150.74	153.48	100.48	197.98	30.64	166.17	59.49	14.25	–[d]

Chemical shifts of the phenyl group (C-1, C-2 and C-6, C-3 and C-5, C-4): [a] 126.16, 127.27, 129.24, 132.06. [b] 126.22, 127.07, 129.30, 131.91. [c] 126.05, 127.21, 129.18, 132.03. [d] 126.16, 127.08, 129.24, 131.85.

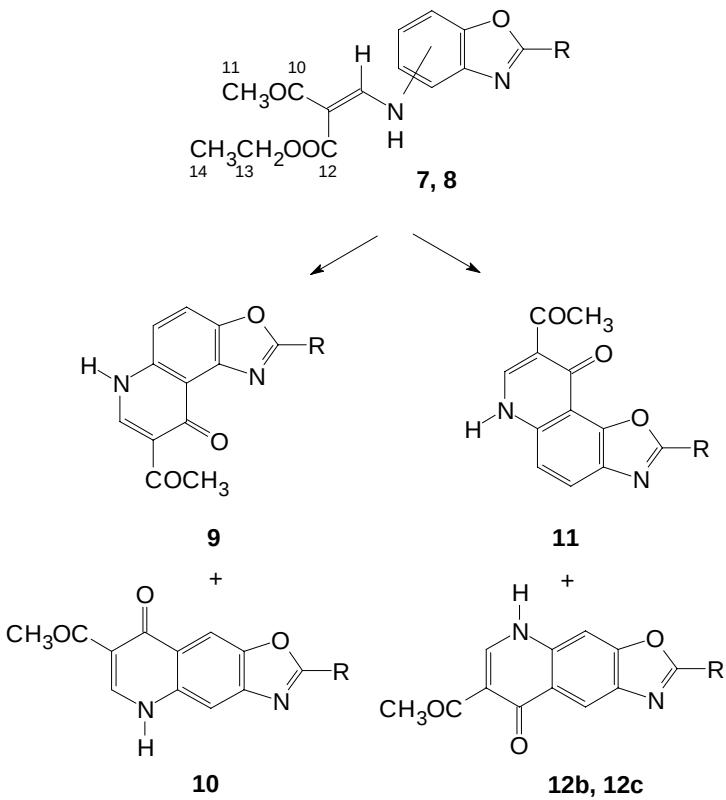
Mass spectra of all the substituted derivatives exhibit the molecular ion. The substitution products **5** and **6** display a peak *m/z* 112 (100%), corresponding to loss of the diacetylethylene fragment. In the case of derivatives **7** and **8** we observed two fragmentation paths. The main fragmentation of compounds **7** consisted in loss of ethyl formate (M – 74), derivatives **8** lost preferentially the acetyl group (M – 43) with subsequent fragmentation. The mass spectral data for derivatives **5**–**8** are given in Table V.

The cyclization of substitution products **7** and **8** takes place at high temperature (250–260 °C) in a mixture of diphenyl ether and biphenyl (Scheme 2). The reaction time was 30–60 min. In most cases the reaction afforded a mixture of angularly and linearly annelated oxazoloquinolones, only the cyclization of compound **8a** gave exclusively the angularly annelated derivative **11a**. The cyclization was also greatly in-

TABLE V
Mass spectra of substitution products **5**–**8**

Compound	*m/z* (relative abundance, %)
5a	244 ($M^{+\bullet}$, 92), 229 (9), 228 (44), 212 (8), 211 (41), 201 (11), 188 (12), 187 (94), 159 (20), 158 (10), 145 (13), 134 (11), 118 (10), 112 (100), 104 (11), 91 (9), 77 (8), 63 (16), 51 (12), 43 (98), 39 (19), 28 (17)
5b	258 ($M^{+\bullet}$, 58), 243 (23), 225 (38), 215 (11), 202 (10), 201 (38), 197 (9), 160 (13), 147 (32), 132 (20), 112 (100), 104 (11), 91 (11), 63 (8), 43 (57), 32 (12), 28 (28)
5c	320 ($M^{+\bullet}$, 49), 305 (59), 287 (30), 277 (12), 263 (22), 235 (15), 209 (50), 160 (20), 132 (26), 112 (100), 104 (24), 91 (16), 77 (22), 63 (20), 43 (85), 39 (9)
6a	244 ($M^{+\bullet}$, 42), 229 (20), 202 (5), 201 (39), 188 (6), 187 (52), 159 (17), 132 (11), 112 (100), 104 (8), 90 (13), 63 (17), 43 (71), 39 (9), 32 (9), 28 (25)
6b	258 ($M^{+\bullet}$, 60), 243 (25), 225 (38), 215 (10), 201 (41), 173 (18), 160 (13), 147 (33), 132 (27), 112 (100), 104 (14), 91 (14), 76 (10), 52 (10), 43 (87), 39 (9), 28 (29)
6c	320 ($M^{+\bullet}$, 58), 305 (20), 287 (27), 263 (18), 210 (20), 209 (65), 166 (10), 132 (22), 112 (100), 104 (19), 103 (12), 91 (10), 77 (19), 63 (20), 43 (56), 39 (9), 28 (19)
7a	274 ($M^{+\bullet}$, 72), 259 (9), 229 (13), 228 (16), 213 (20), 201 (17), 200 (100), 187 (12), 185 (56), 158 (13), 142 (32), 103 (7), 76 (8), 52 (10), 43 (57), 29 (19), 28 (19)
7b	288 ($M^{+\bullet}$, 81), 243 (12), 242 (13), 227 (15), 214 (100), 213 (15), 201 (6), 199 (41), 172 (10), 142 (18), 132 (10), 104 (10), 91 (7), 63 (10), 43 (49), 29 (13), 28 (10)
7c	350 ($M^{+\bullet}$, 56), 289 (10), 282 (7), 277 (18), 276 (87), 274 (51), 261 (51), 200 (100), 185 (49), 158 (18), 142 (49), 104 (12), 91 (14), 51 (19), 43 (80), 29 (15), 28 (30)
8a	274 ($M^{+\bullet}$, 15), 157 (11), 142 (10), 104 (7), 103 (9), 90 (10), 77 (9), 63 (13), 51 (10), 43 (100), 29 (46)
8b	288 ($M^{+\bullet}$, 38), 243 (10), 242 (17), 227 (13), 215 (16), 214 (100), 200 (7), 199 (53), 173 (7), 172 (17), 142 (28), 104 (28), 103 (10), 77 (10), 63 (7), 43 (30), 28 (12)
8c	350 ($M^{+\bullet}$, 10), 104 (13), 103 (23), 91 (5), 77 (8), 63 (33), 51 (19), 43 (100), 29 (17)

fluenced by dilution of the starting solution. The concentration of choice was 1 g of the compound in 50 g of the solvent. Greater concentrations resulted in darkening of the mixture, with lower concentrations the isolation of the products was very difficult. The purity and yields of the cyclocondensation reaction depended also on the purity of the starting substitution products, good yields being achieved only with recrystallized compounds. The obtained cyclic oxazoloquinolones were extraordinarily insoluble and melted above 250 °C. The low solubility made the NMR spectral measurements difficult and, more importantly, made impossible the separation of the angularly and linearly annelated products.



In formulae **7-12** : **a**, R = H
b, R = CH_3
c, R = C_6H_5

SCHEME 2

The analytical data, yields and IR spectra of the cyclocondensation products **9**–**12** are given in Table VI. Except the derivative **11a**, all the products are mixtures of angularly and linearly annelated compounds; the data are given only to characterize the obtained mixtures and not the individual compounds. The ^{1}H NMR spectra (Table VII) of these mixtures of angularly and linearly annelated products show some doubled signals, e.g. those of acetyl group, the proton or methyl group in position 2 of the oxazole nucleus, and in some cases also of the CH–NH protons. The spectra further exhibited characteristic doublets for angularly annelated derivatives **9** (for H-8 and H-9) and **11** (for H-4 and H-5) and singlets, attributable to the H-4 and H-9 protons of the linearly annelated derivatives **10a**–**10c** and **12b**, **12c**. The ratio of the angular to linear isomers was determined by the ^{1}H NMR spectra. Mass spectra of the cyclic products (Table VIII) show the molecular ions. In most cases, the molecules lose preferentially the methyl group.

EXPERIMENTAL

The melting points are uncorrected. Proton and ^{13}C NMR spectra were measured on a Varian VXR-300 instrument (300 MHz for ^{1}H, 75 MHz for ^{13}C) in hexadeuteriodimethyl sulfoxide with tetramethylsilane as internal standard. IR spectra were taken on an M-80 (Zeiss, Jena) spectrometer using the KBr

TABLE VI
Physicochemical properties of cyclocondensation products **9**–**12**

Compound or mixture	Yield, %	Formula M.w.	Calculated/Found			IR spectra	
			% C	% H	% N	$\tilde{\nu}$(CO)	$\tilde{\nu}$(C=C)
9a	22	$C_{12}H_8N_2O_3$	63.16	3.53	12.28	1 581, 1 664	1 543
10a		228.2	63.00	3.50	12.24		
9b	25	$C_{13}H_{10}N_2O_3$	64.46	4.16	11.56	1 581, 1 666	1 539
10b		242.2	64.41	4.11	11.51		
9c	38	$C_{18}H_{12}N_2O_3$	71.05	3.97	9.21	1 624, 1 662	1 541
10c		304.3	71.15	4.02	9.11		
11a	20	$C_{12}H_8N_2O_3$	63.16	3.35	12.28	1 628, 1 666	1 543
		228.2	63.51	3.40	12.32		
11b	20	$C_{13}H_{10}N_2O_3$	64.46	4.16	11.56	1 635, 1 662	1 541
12b		242.2	64.39	4.09	11.49		
11c	33	$C_{18}H_{12}N_2O_3$	71.05	3.97	9.21	1 587, 1 622	1 539
12c		304.3	69.87	3.90	9.18		

TABLE VII
Proton NMR spectra of cyclocondensation products **9**–**12**

Compound	H-4	H-5	H-6	H-7	H-8	H-9	NH	CH_3CO	R
9a[a]	–	–	12.18	–	11.66	11.43	12.33	5.94	–[f]
10a	11.12	–	12.25	–	–	11.84	12.62	6.02	–[f]
9b[b]	–	–	10.08	–	9.15	10.17	10.17	3.57	3.48
10b	9.48	–	10.08	–	–	10.17	10.17	3.77	3.52
9c[c]	–	–	8.40	–	8.23	8.59	8.59	2.52	8.28–8.22
10c	7.85	–	8.40	–	–	8.59	8.59	2.62	7.64–7.58
11a	8.09	7.65	–	8.48	–	–	8.81	2.50	8.92
11b[d]	8.10	7.98	–	8.29	–	8.26	8.48	2.75	2.49
12b	9.30	–	–	8.29	–	–	8.48	2.75	2.49
11c[e]	8.17	7.70	–	8.55	–	–[a]	8.62	2.85	8.28–8.22
12c	9.97	–	–	8.55	–	–	8.62	2.88	7.68–7.55

Population of isomers in the mixture: [a] 3 : 1. [b] 3 : 2. [c] 1 : 1. [d] 2 : 1. [e] 3 : 2. [f] Signal not obvious.

TABLE VIII
Mass spectra of cyclocondensation products **9**–**12**

Compound	*m*/*z* (relative abundance, %)
9a	228 ($M^{+\bullet}$, 77), 214 (13), 213 (100), 185 (8), 157 (5), 145 (34), 130 (21), 129 (5)
10a	103 (5), 102 (9), 75 (15), 63 (7), 62 (8), 53 (15), 52 (8), 43 (30), 28 (10)
9b	242 ($M^{+\bullet}$, 60), 228 (14), 227 (100), 199 (10), 171 (5), 159 (22), 158 (7), 144 (7)
10b	130 (5), 120 (5), 102 (5), 75 (7), 53 (5), 43 (27), 28 (5)
9c	304 ($M^{+\bullet}$, 68), 290 (20), 289 (100), 278 (5), 277 (10), 270 (49), 263 (7)
10c	261 (27), 234 (8), 210 (7), 170 (12), 142 (11), 130 (7), 104 (14), 103 (14), 91 (7), 77 (27), 76 (12), 63 (14), 53 (12), 52 (10), 51 (22), 43 (35), 39 (11)
11a	228 ($M^{+\bullet}$, 32), 214 (5), 213 (47), 171 (13), 170 (100), 169 (13), 154 (59), 153 (18), 152 (13), 145 (12), 142 (34), 140 (49), 115 (18), 77 (35), 76 (12), 63 (12), 51 (42), 50 (14), 43 (10), 39 (19), 28 (7)
11b	242 ($M^{+\bullet}$, 62), 228 (14), 227 (100), 199 (8), 171 (5), 170 (15), 159 (25), 158 (5)
12b	154 (11), 77 (11), 76 (11), 43 (20), 39 (7), 28 (13)
11c	304 ($M^{+\bullet}$, 100), 290 (17), 289 (78), 286 (7), 261 (9), 223 (7), 221 (13), 206 (7)
12c	170 (26), 154 (17), 141 (14), 130 (12), 104 (10), 103 (12), 102 (15), 77 (22), 76 (9), 75 (11), 63 (7), 51 (15), 43 (12), 39 (7)

technique (1 mg in 300 mg KBr). Electronic absorption spectra were recorded on a UV-VIS M-40 spectrophotometer (Zeiss, Jena) in methanol, concentration 1 . 10^{-4} mol dm^{-3}. Mass spectra were obtained with an MS 902S (AEI Kratos) instrument. The nitrobenzoxazoles were prepared according to refs[5,7–11].

Preparation of Aminobenzoxazoles **1** and **2**

A mixture of the nitro compound (0.01 mol), Raney nickel (10% of weight of the nitro derivative) and ethanol (100 ml) was hydrogenated for 24–120 h, the reaction being monitored by TLC and hydrogen consumption. After filtration, the obtained alcoholic solution of the amine was immediately used in the further reaction.

Preparation of Substitution Derivatives **5**–**8**

The alcoholic solution of the amine, obtained in the above experiment, was mixed with 3-ethoxymethylene-2,4-pentanedione (**3**) or ethyl 2-ethoxymethylene-3-oxobutanoate (**4**) (0.01 mol-equivalent). The mixture was refluxed for 15–60 min and the reaction was monitored by TLC. The reaction mixture was concentrated in vacuo on a rotatory evaporator and the product was crystallized from an appropriate solvent. The yields, reaction times, melting points and crystallization solvents are given in Table I, their IR and UV spectra in Table II, ^{1}H NMR spectra in Table III, ^{13}C NMR spectra in Table IV and mass spectra in Table V.

Preparation of Cyclocondensation Products **9**–**12**

A mixture of the substitution product **7** or **8** (1 g) and Dowtherm (50 g) was heated at 250–260 °C for 30–60 min. In most cases, cooling of the mixture afforded a gelatinous mass which was washed with hexane and ether to remove the residual solvent. The physicochemical data for compounds **9**–**12** and their IR spectra are given in Table VI, their ^{1}H NMR spectra in Table VII and mass spectra in Table VIII.

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