SYNTHESIS OF SOME DIAROYLHYDRAZINES, 1,3,4-OXADIAZOLES AND PYRIDAZIN-3-ONES BEARING THIOPHENE NUCLEUS

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Diaroylhydrazines *I* (refs[1,2]) were used for the preparation of 1,3,4-oxadiazoles *II* (refs[3,4]) and pyridazinones *III* (ref.[5]), respectively.

EXPERIMENTAL

All melting points are uncorrected. The IR spectra were measured on Perkin–Elmer Spectrophotometer 297 using KBr wafer technique. The electronic spectra were done on Perkin–Elmer UV spectrophotometer 505 using ethanolic solutions. Analyses were carried out by the Micro-analytical unit, Cairo University, and Organic Analysis Lab., N. R. C., Dokki, Cairo, Egypt. For the physical and spectral data cf. Tables I and II.

1-Aroyl-2-[α-aracyl-β-(2-thienyl)]acroylhydrazines (*Ia – Ie*)

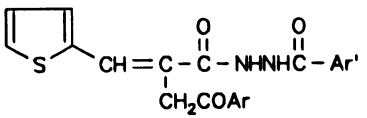
To a solution of the hydrazide[1,2] (0.01 mol) in 50 ml dry benzene, the acyl chloride (0.01 mol) was added. The reaction mixture was heated under reflux for 2 h. The solvent was distilled off under reduced pressure. The yellow solid obtained was washed thoroughly with water, drained, and recrystallized from methanol.

2-Aryl-5-[α-aracyl-β-(2-thienyl)]acryl-1,3,4-oxadiazoles (*IIa – IIl*)

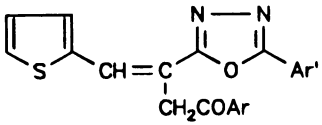
Phosphorus oxychloride (10 ml) was added dropwise to 1.0 g of the diaroylhydrazine *I*. The reaction mixture was refluxed for 20 min left to cool, and poured onto crushed ice. The yellowish brown solid precipitated was filtered off, washed with water and recrystallized from benzene–petroleum ether mixture to produce compound *II*.

1-Benzoyl-6-aryl-4-thienylidene-1,6-dihydropyridazin-3-(2*H*)ones (*IIIa – IIId*)

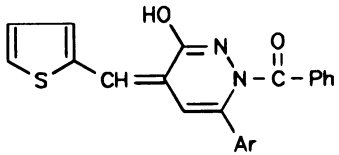
To the diaroylhydrazine *I* (1.0 g) 30 ml of HCl–CH_3COOH mixture (1 : 1) was added. The reaction mixture was heated under reflux for 1 h and then left to cool. The pale yellow solid formed was filtered off, washed with water and finally recrystallized from ethanol to give compound *III*.



I



II



III

In formulae *I* – *III* :

	Ar	Ar'
a	C_6H_5	C_6H_5
b	4-$CH_3C_6H_4$	C_6H_5
c	4-$CH_3OC_6H_4$	C_6H_5
d	4-ClC_6H_4	C_6H_5
e	C_6H_5	4-$CH_3C_6H_4$
f	4-$CH_3C_6H_4$	4-$CH_3C_6H_4$
g	4-$CH_3OC_6H_4$	4-$CH_3C_6H_4$
h	4-ClC_6H_4	4-$CH_3C_6H_4$
i	C_6H_5	4-$CH_3OC_6H_4$
j	4-$CH_3C_6H_4$	4-$CH_3OC_6H_4$
k	4-$CH_3OC_6H_4$	4-$CH_3OC_6H_4$
l	4-ClC_6H_4	4-$CH_3OC_6H_4$

TABLE I
Physical data for synthesized compounds

Compound	Formula (M. w.)	M. p., °C Yield, %	Calculated/Found			
			% C	% H	% N	% S
Ia	$C_{22}H_{18}N_2O_3S$	195	67.69	4.61	7.17	8.20
	(390)	80	67.60	4.80	7.30	8.80
Ib	$C_{23}H_{20}N_2O_3S$	204	68.31	4.95	6.93	–
	(404)	80	68.70	5.50	6.99	–
Ic	$C_{23}H_{20}N_2O_4S$	213	65.71	4.76	6.66	–
	(420)	75	65.90	4.50	7.20	–
Id	$C_{22}H_{17}ClN_2O_3S$	231	–	–	–	7.53
	(424.5)	78	–	–	–	7.80
Ie	$C_{23}H_{20}N_2O_3S$	235 – 237	68.83	4.95	–	–
	(404)	83	68.40	5.30	–	–
If	$C_{24}H_{22}N_2O_3S$	254 – 256	68.89	5.26	–	–
	(418)	88	68.70	5.70	–	–
Ig	$C_{24}H_{22}N_2O_4S$	244	66.35	5.06	6.46	–
	(434)	85	66.50	4.70	6.60	–
Ih	$C_{23}H_{19}ClN_2O_3S$	253	62.94	4.33	–	–
	(438.5)	78	63.10	4.00	–	–
Ii	$C_{23}H_{20}N_2O_4S$	223	65.71	4.76	–	–
	(420)	80	65.30	4.40	–	–
Ij	$C_{24}H_{22}N_2O_4S$	229	66.35	5.06	–	–
	(434)	80	66.30	5.00	–	–
Ik	$C_{24}H_{22}N_2O_5S$	235	64.00	4.88	–	–
	(450)	80	64.50	5.20	–	–
Il	$C_{23}H_{19}ClN_2O_4S$	240	–	–	6.16	7.04
	(454.5)	78	–	–	5.60	6.40
IIa	$C_{22}H_{16}N_2O_2S$	160	70.96	4.30	7.52	–
	(372)	70	70.50	3.90	7.00	–
IIb	$C_{23}H_{18}N_2O_2S$	170	71.50	4.66	–	8.29
	(386)	75	71.20	5.10	–	8.20
IIc	$C_{23}H_{18}N_2O_3S$	189	68.65	4.47	–	–
	(402)	73	68.90	4.70	–	–
IId	$C_{22}H_{15}ClN_2O_2S$	204	64.94	3.69	–	7.83
	(406.5)	78	64.80	4.10	–	7.90

TABLE I
(*Continued*)

Compound	Formula (M. w.)	M. p., °C Yield, %	Calculated/Found			
			% C	% H	% N	% S
IIe	$C_{23}H_{18}N_2O_2S$	133 – 134	71.50	4.66	–	–
	(386)	81	71.60	4.80	–	–
IIf	$C_{24}H_{20}N_2O_2S$	139	72.00	5.00	–	–
	(400)	74	71.90	5.10	–	–
IIg	$C_{24}H_{20}N_2O_3S$	152	69.23	4.80	–	–
	(416)	75	69.00	5.40	–	–
IIh	$C_{23}H_{17}ClN_2O_2S$	213	65.63	4.04	–	7.60
	(404)	76	65.60	4.50	–	7.60
IIi	$C_{23}H_{18}N_2O_3S$	145 – 147	68.65	4.47	–	–
	(402)	82	68.00	4.60	–	–
IIj	$C_{24}H_{20}N_2O_3S$	112 – 114	69.23	4.80	–	–
	(416)	78	69.70	4.30	–	–
IIk	$C_{24}H_{20}N_2O_4S$	144	66.66	4.63	–	–
	(432)	80	66.40	5.20	–	–
III	$C_{23}H_{17}ClN_2O_3S$	175	63.23	3.89	–	7.33
	(436.5)	77	63.70	3.30	–	7.90
IIIa	$C_{22}H_{16}N_2O_2S$	180	70.96	4.30	7.52	–
	(372)	73	71.30	4.70	8.09	–
IIIb	$C_{23}H_{18}N_2O_2S$	186	71.50	4.66	7.25	–
	(368)	75	71.20	4.60	8.07	–
IIIc	$C_{23}H_{18}N_2O_3S$	198	68.65	4.47	6.96	–
	(402)	77	68.50	5.00	7.20	–
IIId	$C_{22}H_{15}ClN_2O_2S$	212	–	–	–	7.87
	(606.5)	71	–	–	–	8.00

TABLE II
Spectral data for synthesized compounds

Compound	IR spectra[a]			UV spectra λ_{max} ($\varepsilon_{max} \cdot 10^{-4}$)
	ν(NH)	ν(C=O)	ν(–C=N–N=C–)	
Ia	3 150 – 3 290	1 660		201 (7.31), 223 (4.60), 263 (4.00), 339 (3.12), 418 (4.20)
Ib	3 150 – 3 250	1 670		202 (8.22), 223 (5.11), 270 (5.00), 373 (3.45), 423 (4.92)
Ic	3 150 – 3 200	1 645		202 (7.90), 223 (4.80), 262 (3.90), 336 (3.00), 420 (4.00)
Id	3 150 – 3 300	1 650		203 (6.60), 221 (4.03), 270 (4.23), 340 (2.80), 420 (4.00)
Ie	3 150 – 3 280	1 660		205 (8.00), 238 (4.99), 270 (5.80), 337 (3.00), 420 (4.51)
If	3 150 – 3 280	1 670		205 (8.50), 237 (5.05), 270 (6.00), 338 (3.20), 420 (4.00)
Ig	3 150 – 3 240	1 660		203 (8.20), 238 (5.48), 270 (6.52), 338 (3.23), 420 (5.30)
Ih	3 150 – 3 250	1 640		202 (6.52), 225 (5.02), 270 (6.03), 335 (2.02), 423 (3.00)
Ii	3 150 – 3 240	1 660		205 (10.5), 238 (7.61), 263 (6.30), 338 (4.80), 425 (6.30)
Ij	3 150 – 3 200	1 650		205 (8.60), 237 (4.34), 260 (3.85), 338 (2.89), 420 (3.90)
Ik	3 100 – 3 200	1 640		204 (6.70), 237 (3.26), 270 (4.00), 343 (2.25), 423 (3.10)
Il	3 150 – 3 240	1 650		204 (8.97), 237 (3.29), 270 (3.85), 340 (2.27), 420 (2.75)
IIa		1 690	1 595, 1 610	204 (7.44), 236 (5.60), 266 (4.69), 336 (3.60), 424 (4.65)
IIb		1 690	1 590, 1 610	203 (7.87), 233 (6.05), 269 (5.30), 338 (3.37), 422 (4.34)
IIc		1 690	1 590, 1 610	203 (6.80), 235 (5.52), 265 (4.80), 335 (3.16), 423 (4.27)
IId		1 690	1 590, 1 610	203 (6.40), 229 (5.30), 267 (4.07), 337 (2.99), 428 (4.80)
IIe		1 690	1 590, 1 616	204 (7.74), 237 (6.00), 268 (4.50), 338 (2.00), 423 (2.25)

TABLE II
(*Continued*)

Compound	IR spectra[a]			U V spectra λ_{max} ($\varepsilon_{max} \cdot 10^{-4}$)
	ν(NH)	ν(C=O)	ν(–C=N–N=C–)	
IIf		1 695	1 595, 1 610	206 (9.16), 239 (5.60), 268 (4.87), 338 (2.73), 425 (2.94)
IIg		1 690	1 590, 1 610	205 (8.30), 238 (6.01), 267 (5.58), 337 (2.77), 422 (2.40)
IIh		1 690	1 590, 1 610	203 (9.01), 237 (4.90), 265 (4.05), 338 (2.63), 425 (2.73)
IIi		1 690	1 590, 1 610	205 (7.43), 224 (3.51), 259 (5.23), 338 (2.51), 423 (3.20)
IIj		1 690	1 590, 1 610	204 (5.20), 223 (3.45), 258 (4.78), 340 (2.08), 422 (2.80)
IIk		1 690	1 590, 1 610	204 (5.90), 223 (3.30), 265 (4.50), 339 (2.00), 423 (2.15)
IIl		1 690	1 590, 1 610	205 (5.67), 223 (3.45), 260 (4.63), 338 (2.30), 424 (2.91)
IIIa	2 700 – 3 200	1 660	1 610[b]	205 (8.13), 235 (9.76), 310 (1.16)
IIIb	2 650 – 3 200	1 660	1 610[b]	205 (7.23), 235 (7.79), 310 (1.20)
IIIc	2 650 – 3 250	1 660	1 610[b]	204 (7.76), 237 (7.78), 312 (1.25)
IIId	2 700 – 3 250	1 660	1 610[b]	202 (6.51), 236 (5.76), 310 (1.30)

[a] Wavenumber in cm^{-1}; [b] ν(C≡N).

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