5-ARYL-2-FURANCARBALDEHYDE HYDRAZONES AND RELATED COMPOUND

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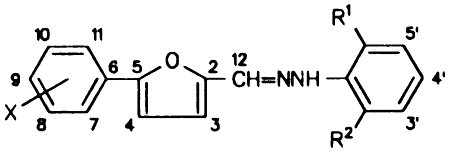
A series of 5-aryl-2-furancarbaldehyde 2,6-dialkylphenylhydrazones (*Ia* – *Iv*) and dimethylhydrazones (*IIa* – *IIf*) as well as related compound benzo[*b*]furan-2-carbaldehyde dimethylhydrazone (*III*) were prepared. Compounds *Ia* – *Iv* were synthesized by condensing 5-aryl-2-furancarbaldehydes with 2,6-dialkylphenylhydrazines, compounds *IIa* – *IIf* were obtained from the same starting compounds and *N,N*-dimethylhydrazine. The intermediate 5-aryl-2-furancarbaldehydes were prepared by reaction of aryldiazonium chlorides with 2-furancarbaldehyde. The structure of the compounds were proven by ^{1}H, ^{13}C and ^{15}N NMR spectra.

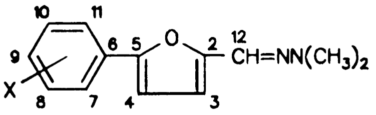
2,6-Dialkylphenylhydrazines[1,2], especially 2,6-dimethylphenylhydrazine were used for the synthesis of fungicidally active compounds[3 – 5]. The latter was employed[6] for an interesting synthesis of 3-amino-1,3-thiazolidinediones, hypnotically active thiazolines[7,8] and 5-pyrazolecarboxylic acid derivatives having a sedative, hypnotic and myorelaxation effects. 2,6-Dimethylphenylhydrazinium chloride was reported[9] to react with phenyl ketones to yield substituted pyrazoles utilizable as scintillators and fluorescent bleachers avoiding yellowing of synthetic fibres.

2,6-Dialkylphenylhydrazines were used for preparation of indolylformazanes, indolylazetidinones, tetrazolium salts[10] and 1,2,4-triazole derivatives[11]. Also new rearrangements of 2,6-dialkylphenylhydrazones were published[2,12] and an action of mild oxidation reagents on 2,6-dimethylphenylhydrazine and its condensation products was investigated[13 – 15]. Our previous papers concerned 2,6-dialkylphenylhydrazine reactions with various kind of electrophilic agents[16,17].

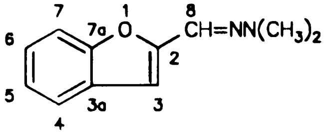
This paper describes reactions of 5-aryl-2-furancarbaldehydes with 2,6-dialkylphenylhydrazines to give hydrazones *Ia* – *Iv* in excellent yields. 5-Aryl-2-furancarbaldehyde dimethylhydrazones (*IIa* – *IIf*) were readily prepared from 5-aryl-2-furancarbaldehydes and *N,N*-dimethylhydrazine in refluxing toluene, with catalytic amount of 4-methylbenzenesulfonic acid. The compound *III* was prepared analogously



	R^1	R^2	X
Ia	CH_3	CH_3	H
Ib	CH_3	C_2H_5	H
Ic	C_2H_5	C_2H_5	H
Id	CH_3	CH_3	4-CH_3
Ie	CH_3	C_2H_5	4-CH_3
If	C_2H_5	C_2H_5	4-CH_3
Ig	CH_3	CH_3	2-NO_2
Ih	CH_3	C_2H_5	2-NO_2
Ii	C_2H_5	C_2H_5	2-NO_2
Ij	CH_3	CH_3	3-NO_2
Ik	CH_3	C_2H_5	3-NO_2
Im	C_2H_5	C_2H_5	3-NO_2
In	CH_3	CH_3	4-NO_2
Io	CH_3	C_2H_5	4-NO_2
Ip	C_2H_5	C_2H_5	4-NO_2
Iq	CH_3	CH_3	4-Cl
Ir	CH_3	C_2H_5	4-Cl
Is	C_2H_5	C_2H_5	4-Cl
It	CH_3	CH_3	3,4-Cl_2
Iu	CH_3	C_2H_5	3,4-Cl_2
Iv	C_2H_5	C_2H_5	3,4-Cl_2



	X
IIa	H
IIb	4-CH_3
IIc	3-NO_2
IId	4-NO_2
IIe	4-Cl
IIf	3,4-Cl_2



III

TABLE I
Characteristic data of compounds *Ia* – *Iv*, *IIa* – *IIf* and *III*

Compound	Formula (M. w.)	M. p., °C Yield, %	Calculated/Found			UV spectra λ_{max} (log ε)	
			% C	% H	% N		
Ia	$C_{19}H_{18}N_2O$	138 – 140	78.59	6.25	9.65	342	
	(290.4)	75	78.42	6.36	9.72	(3.49)	
Ib	$C_{20}H_{20}N_2O$	86 – 87	78.92	6.62	9.20	341	
	(304.4)	69	78.88	6.48	9.32	(3.36)	
Ic	$C_{21}H_{22}N_2O$	102 – 106	79.21	6.96	8.80	342	
	(318.4)	73	79.31	6.91	8.85	(3.49)	
Id	$C_{20}H_{20}N_2O$	155 – 158	78.92	6.62	9.20	344	276
	(304.4)	89	78.84	6.49	9.10	(3.46)	(3.06)
Ie	$C_{21}H_{22}N_2O$	156 – 158	79.21	6.96	8.80	343	277
	(318.4)	63	79.22	6.90	8.72	(3.36)	(3.04)
If	$C_{22}H_{24}N_2O$	141 – 143	79.48	7.28	8.43	344	260
	(332.4)	72	79.42	7.02	8.40	(3.57)	(3.03)
Ig	$C_{19}H_{17}N_3O_3$	127 – 130	68.05	5.11	12.53	333	238
	(335.4)	82	68.90	5.23	12.64	(3.28)	(3.26)
Ih	$C_{20}H_{19}N_3O_3$	72 – 74	68.75	5.48	12.03	333	243
	(349.4)	58	68.62	5.42	12.09	(3.23)	(3.19)
Ii	$C_{21}H_{21}N_3O_3$	102 – 104	69.41	5.82	11.56	334	238
	(363.4)	73	69.45	5.69	11.45	(3.31)	(3.19)
Ij	$C_{19}H_{17}N_3O_3$	100 – 103	68.05	5.11	12.53	349	259
	(335.4)	91	68.40	5.01	12.29	(3.38)	(3.18)
Ik	$C_{20}H_{19}N_3O_3$	103 – 106	68.75	5.48	12.03	350	258
	(349.4)	64	68.59	5.62	12.09	(3.50)	(3.24)
Im	$C_{21}H_{21}N_3O_3$	128 – 130	69.41	5.82	11.56	350	257
	(363.4)	71	69.56	5.63	11.72	(3.46)	(3.18)
In	$C_{19}H_{17}N_3O_3$	198 – 201	68.05	5.11	12.53	413	312
	(335.4)	93	68.24	5.18	12.49	(3.35)	(3.19)
Io	$C_{20}H_{19}N_3O_3$	151 – 155	68.75	5.48	12.03	414	312
	(349.4)	89	68.66	5.38	12.23	(3.38)	(3.20)
Ip	$C_{21}H_{21}N_3O_3$	143 – 147	69.41	5.82	11.56	415	312
	(363.4)	92	69.45	5.89	11.66	(3.28)	(3.19)
Iq	$C_{19}H_{17}ClN_2O$	144 – 146	70.26	5.28	8.62	349	
	(324.8)	91	70.34	5.25	8.64	(3.51)	

TABLE I
(*Continued*)

Compound	Formula (M. w.)	M. p., °C Yield, %	Calculated/Found			UV spectra λ_{max} (log ε)	
			% C	% H	% N		
Ir	$C_{20}H_{19}ClN_2O$	143 – 145	70.90	5.65	8.27	349	
	(338.8)	89	70.79	5.80	8.34	(3.52)	
Is	$C_{21}H_{21}ClN_2O$	117 – 118	71.48	6.00	7.94	350	
	(352.8)	96	71.44	6.12	7.81	(3.53)	
It	$C_{19}H_{16}Cl_2N_2O$	120 – 122	63.52	4.49	7.80	352	
	(359.2)	84	63.38	4.52	7.77	(3.48)	
Iu	$C_{20}H_{18}Cl_2N_2O$	117 – 118	64.35	4.86	7.50	353	
	(373.3)	80	64.28	4.72	7.68	(3.49)	
Iv	$C_{21}H_{20}Cl_2N_2O$	107 – 109	65.12	5.20	7.23	354	
	(387.3)	81	65.32	5.16	7.12	(3.51)	
IIa	$C_{13}H_{14}N_2O$	[a]	72.87	6.59	13.07	340	
	(214.3)	79	72.66	6.34	13.24	(3.54)	
IIb	$C_{14}H_{16}N_2O$	66 – 67	73.66	7.06	12.27	341	261
	(228.3)	69	73.42	7.26	12.34	(3.52)	(3.03)
IIc	$C_{13}H_{13}N_3O_3$	88 – 90	60.23	5.05	16.21	346	258
	(259.3)	75	60.13	5.12	16.45	(3.39)	(3.15)
IId	$C_{13}H_{13}N_3O_3$	147 – 149	60.23	5.05	16.21	414	307
	(259.3)	84	60.03	5.21	16.46	(3.33)	(3.13)
IIe	$C_{13}H_{13}ClN_2O$	80 – 83	62.78	5.27	11.26	346	264
	(248.7)	78	62.81	5.17	11.38	(3.51)	(3.06)
IIf	$C_{13}H_{12}Cl_2N_2O$	96 – 97	55.14	4.27	9.89	352	268
	(283.1)	80	55.01	4.14	9.69	(3.50)	(3.08)
III	$C_{11}H_{12}N_2O$	42 – 44	70.19	6.43	14.88	324	225
	(188.2)	92	70.26	6.32	14.72	(3.54)	(2.90)

[a] B. p. = 120 – 121 °C/26.6 Pa.

starting from benzo[*b*]furan-2-carbaldehyde. Characteristic data and UV spectra of synthesized compounds are listed in Table I.

The structure of the studied compounds has been confirmed by ^1H and ^{13}C NMR spectroscopy (Tables II – V). The ^1H NMR spectra display doublets of H-3 and H-4 protons of furan ring with coupling constant $J(3,4)$ = 3.5 Hz and a singlet of H-12. An assignment of aromatic protons was done on the basis of characteristic splitting and the substituent chemical shift values for substituted benzenes[18].

^{13}C NMR spectra were measured for selected compounds of the series *I* ($R^1 = R^2 = CH_3$) to found out the influence of X-substituents attached at the benzene ring. For the assignment of carbon atoms the selective heteronuclear decoupling and semiselective INEPT experiment[19] was applied. Carbon atoms of benzo[*b*]furan skeleton in the compound *III* were assigned according to analogy of chemical shift values with those of benzo[*b*]furan and its derivatives[20]. The ^1H and ^{13}C NMR data of *III* are given in experimental part.

The chemical shifts of H-12 and C-12 are almost unaffected by X substituents at the benzene ring. Only the chemical shifts of H-4 and C-4 respectively depend on the character and position of substituents.

In order to determine the configuration of the substituents on the double bond of the prepared hydrazones the ^{15}N NMR spectra of *Iq* and *IIe* were measured.

The ^{15}N chemical shift for CH=N nitrogen is $\delta = -40.00$ in *Iq* and $\delta = -22.5$ in *IIe* and for CH=N–NH– nitrogen $\delta = -244.00$ in *Iq* and -268.00 in *IIe*. $J(^{15}N,^1H)$ coupling constants were determined for *Iq*: ^2J(N,H-12) = 3.45 and ^3J(N,H-12) = 6.80 Hz. The value 3.45 Hz of geminal N,H coupling confirms the *E*-isomer of this compound, which is in agreement with coupling constant values of hydrazones in ref.[21].

These compounds were tested as potential pesticides by standard methods for fungicide[22] and herbicide[23] activities, but none of them achieved properties of the references.

EXPERIMENTAL

Melting points were determined on Kofler hot plate apparatus and were uncorrected. All NMR experiments were performed on a Bruker AM-300 FT NMR spectrometer at 298 K, solvent $(CD_3)_2CO$. ^1H NMR spectra were recorded at 300 MHz. The following measurement conditions were used for ^{13}C NMR spectra: 75.43 MHz, tube size 10 mm, standard internal TMS, pulse width 12 μs, flip angle 50°, acquisition time 0.4 s, pulse delay 2 s, spectral width 5 KHz. For the signal assignment the selective SF decoupling and semiselective INEPT experiment was carried out, with the optimalization for *J*(long-range) = 7 Hz. Results are given in δ, ppm.

^1H decoupled ^{15}N spectrum was measured using the inverse gated pulse sequence (without NOE) at frequency 30.42 Hz. $Cr(acac)_3$ was used to reduce the relaxation time for ^{15}N nucleus. The chemical shifts are referenced to CH_3NO_2. For the estimation of coupling constants ^2J(N,H) and ^3J(N,H) a ^1H-coupled INEPT pulse sequence was used with semiselective polarization transfer from ^1H to ^{15}N, via long-range and selective proton decoupling[24]. Proton H-12 was used for the polarization transfer

TABLE II
^{1}H NMR spectra of compounds *Ia* – *Iv*

Compound	H-12	H-3[a]	H-4	NH	R^1		R^2		Other signals[b,c]
Ia	7.55 s	6.58 d	6.88 d	8.48 bs	2.33 s		2.33 s		7.71 dd (H-7,11), 7.27 tt (H-9), 7.40 t (H-8,10)
Ib	7.51 s	6.56 d	6.87 d	8.44 bs	2.31 s		2.74 q	1.22 t	
Ic	7.51 s	6.55 d	6.87 d	8.24 bs	2.72 q	1.22 t	2.72 q	1.22 t	
Id	7.52 s	6.54 d	6.80 d	8.45 bs	2.33 s		2.33 s		7.61 d (H-7,11), 7.19 d (H-8,10)
Ie	7.51 s	6.53 d	6.80 d	8.40 bs	2.32 s		2.73 q	1.22 t	
If	7.50 s	6.52 d	6.80 d	8.38 bs	2.71 q	1.22 t	2.71 q	1.22 t	
Ig	7.50 s	6.63 d	6.83 d	8.58 bs	2.32 s		2.32 s		7.86 dd (H-8), 7.69 dt (H-9), 7.54 dt (H-10), 7.78 (H-11)
Ih	7.47 s	6.62 d	6.83 d	8.58 bs	2.31 s		2.72 q	1.20 t	
Ii	7.45 s	6.61 d	6.82 d	8.58 bs	2.70 q	1.20 t	2.70 q	1.20 t	
Ij	7.52 s	6.64 d	7.18 d	8.62 bs	2.34 s		2.34 s		8.50 t (H-7), 8.11 m (H-9,11), 7.69 dd (H-10)
Ik	7.51 s	6.62 d	7.16 d	8.59 bs	2.32 s		2.74 q	1.24 t	
Im	7.50 s	6.61 d	7.17 d	8.58 bs	2.71 q	1.23 t	2.71 q	1.23 t	
In	7.52 s	6.67 d	7.23 d	8.70 bs	2.33 s		2.33 s		7.94 d (H-7,11), 8.27 d (H-8,10)
Io	7.50 s	6.66 d	7.22 d	8.67 bs	2.32 s		2.73 q	1.22 t	
Ip	7.49 s	6.64 d	7.22 d	8.67 bs	2.71 q	1.22 t	2.71 q	1.22 t	
Iq	7.52 s	6.55 d	6.92 d	8.53 bs	2.33 s		2.33 s		7.41 d (H-7,11), 7.72 d (H-8,10)
Ir	7.49 s	6.54 d	6.88 d	8.50 bs	2.33 s		2.75 q	1.22 t	
Is	7.49 s	6.54 d	6.90 d	8.50 bs	2.70 q	1.18 t	2.70 q	1.18 t	
It	7.50 s	6.58 d	7.03 d	8.57 bs	2.31 s		2.31 s		7.86 d (H-7), 7.58 d (H-10), 7.69 dd (H-11)
Iu	7.50 s	6.59 d	7.04 d	8.57 bs	2.31 s		2.80 q	1.22 t	
Iv	7.48 s	6.56 d	7.02 d	8.55 bs	2.80 q	1.22 t	2.80 q	1.22 t	

[a] *J*(3,4) = 3.5 Hz; [b] the data are given for derivatives, where R^1 = R^2 = CH$_3$, the values for other corresponding derivatives are nearly the same; [c] for all compounds 6.95 – 7.10 m (H-3',4',5').

and NH for the selective decoupling. The experiment was optimized for $J(N,H) = 4$ Hz. The spectral width for both experiments was 9 433.96 Hz, 32 K data points, digital resolution 0.57 Hz per point.

UV spectra were measured on a M-40 (Zeiss, Jena) spectrophotometer in methanol (λ_{max}/log ε; λ_{max} in nm, ε in $m^2 mol^{-1}$).

The starting compounds were prepared: 2,6-dialkylphenylhydrazinium chlorides (refs[1,2]), (the corresponding hydrazines were liberated with sodium hydroxide); 5-aryl-2-furancarbaldehydes (ref.[25]) and benzo[*b*]furan-2-carbaldehyde (ref.[26]).

5-Phenyl-3-furancarbaldehyde 2,6-Dimethylphenylhydrazone (*Ia*)

2,6-Dimethylphenyl hydrazine (2.7 g, 20 mmol) in toluene (10 ml) was added to solution of 5-phenyl-2-furancarbaldehyde (3.44 g, 20 mmol) in toluene (30 ml) and a catalytic amount of acetic acid (0.5 ml). The mixture was refluxed for 2 h, the solvent was distilled off under reduced pressure and residue was crystallized from ethanol.

This procedure was also applied for preparation of the compounds *Ib* – *Iv*. Characteristic data and UV spectra for compounds *Ia* – *Iv* are listed in Table I.

5-Phenyl-2-furancarbaldehyde Dimethylhydrazone (*IIa*)

A stirred solution of distilled 5-phenyl-2-furancarbaldehyde (1.72 g, 10 mmol) in toluene (10 ml) containing a catalytic amount of 4-methylbenzenesulfonic acid (5 mg) was treated carefully with *N*,*N*-dimethylhydrazine (0.60 g, 10 mmol). The solution was then refluxed for 2 h and water formed during the reaction was removed in Dean–Stark trap. The solvent was removed under reduced pressure and residue was distilled in vacuo.

TABLE III
^{1}H NMR spectra of compounds *IIa* – *IIf*

Compound	H-12	H-3[a]	H-4	CH_3[b]	Other signals
IIa	7.19 s	6.51 d	6.84 d	2.93 s	7.70 dd (H-7,11), 7.26 tt (H-9), 7.41 t (H-8,10)
IIb[c]	7.21 s	6.51 d	6.80 d	2.95 s	7.62 d (H-7,11), 2.34 (CH_3), 7.22 d (H-8,10)
IIc	7.21 s	6.55 d	7.12 d	3.00 s	8.47 t (H-7), 8.10 m (H-9,11) 7.66 t (H-10)
IId	7.21 s	6.59 d	7.21 d	3.03 s	7.94 d (H-7,11), 8.27 d (H-8,10)
IIe	7.19 s	6.51 d	6.91 d	2.95 s	7.42 d (H-7,11), 7.72 d (H-8,10)
IIf	7.18 s	6.52 d	7.00 d	2.96 s	7.82 d (H-7), 7.52 d (H-10), 7.62 dd (H-11)

[a] $J(3,4) = 3.5$ Hz; [b] $N(CH_3)_2$; [c] 2.32 s ($C9-CH_3$).

TABLE IV

^{13}C NMR spectra of some selected compounds of the series *I* ($R^1 = R^2 = CH_3$)

Compound	C-2	C-3	C-4	C-5	C-6	C-7	C-8	C-9	C-10	C-11	C-12	CH_3[a]
Ia[b]	153.70	110.21	108.09	152.52	131.40	124.23	129.54	128.10	129.54	124.23	125.23	18.94
Id	154.00	110.27	107.33	152.12	128.84	124.20	129.51	137.90	129.51	124.20	126.37	18.96[c]
Ig	154.35	109.54	112.41	148.13	132.10	140.26	129.12	132.80	129.27	124.62	125.30	18.96
Ij	153.89	110.18	110.65	151.13	132.97	118.36	149.75	122.14	131.04	129.82	125.03	18.90
In[d]	154.83	112.60	110.48	151.39	137.04	124.09	124.52	147.00	124.52	124.09	125.10	18.86
Iq	152.91	110.23	108.85	152.50	130.20	125.70	129.66	133.10	129.66	125.70	125.37	18.91
It	153.56	110.16	110.16	151.03	133.20	125.86	132.63	130.77	131.81	123.92	125.49	18.87

[a] $R^1 = R^2 = CH_3$; [b] 140.60 (C-1'), 132.30(C-2'), 129.54(C-3'), 125.80(C-4'), 18.91(CH_3); [c] 21.20 (C9–CH_3); [d] 140.00(C-1'), 132.79(C-2'), 129.54(C-3'), 125.70(C-4'), 18.80(CH_3).

TABLE V

^{13}C NMR spectra of compounds *IIa* – *IIf*

Compound	C-2	C-3	C-4	C-5	C-6	C-7	C-8	C-9	C-10	C-11	C-12	CH_3[a]
IIa	155.21	108.64	108.32	151.10	131.66	124.45	129.58	128.01	129.58	124.45	121.82	42.68
IIb	153.45	108.85	107.28	153.28	129.07	124.19	130.16	137.68	130.16	124.19	123.21	42.73[b]
IIc	154.98	110.58	108.61	150.52	133.15	118.19	149.74	121.98	130.87	129.66	121.83	42.64
IId	156.00	112.71	109.04	151.02	137.24	124.29	125.10	146.80	125.10	124.29	121.49	42.59
IIe	154.07	108.86	108.86	151.89	130.45	125.63	129.68	132.87	129.68	125.63	122.62	42.67
IIf	154.61	106.78	110.18	150.34	133.11	125.45	131.95	130.40	131.67	123.73	122.11	42.62

[a] $N(CH_3)_2$; [b] 21.19(C9–CH_3).

Compounds *IIb* – *IIf* and *III* were prepared in the same manner and were crystallized from ethanol. Characteristic data and UV spectra of *IIa* – *IIf* and *III* are listed in Table I.

NMR spectra of *III*: ^{1}H: 7.25 bs, 1 H (H-8); 6.77 d, 1 H (H-3), $J(3,8)$ = 0.8 Hz); 3.01 s, 6 H (CH_3); 7.15 – 7.55 m, (H-4,5,6,7). ^{13}C: 156.12 (C-2), 102.51(C-3), 129.98(C-3a), 121.32(C-4), 123.60(C-5), 124.58(C-6), 111.40(C-7), 155.24(C-7a), 121.40(C-8), 42.52(CH_3).

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