REACTIONS AND SPECTRAL PROPERTIES OF 2-AMINO-3-CYANO-4,5-DISUBSTITUTED FURANE DERIVATIVES

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Propanedinitrile reacts with acyloins *I*—*III* to give 2-amino-3-cyano-4,5-disubstituted furanes *IV*—*VI*; the latter react with formamide to give 4-amino-5,6-disubstituted furo[2,3-*b*]pyrimidines *VII*—*IX*. The derivative *IV* reacts with acetanhydride to 2-acetylamino-3-cyano-4,5-di(2-furyl)furane (*X*), and aminofurane *VI* reacts with acetanhydride to give 2-(N,N-diacetylamino)-3-cyano-4,5-diphenylfurane (*XI*). With aldehydes the aminofuranes *IV*—*VI* form the corresponding azomethines *XII*—*XIV*. The synthetized derivatives were identified by IR, UV, ^{1}H-NMR and mass spectra.

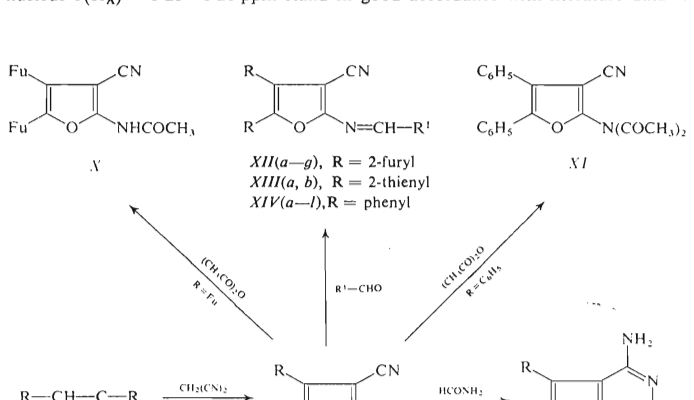
The principal methods of preparation of aminofurane derivatives are: *a*) reduction of nitrofurane derivatives; *b*) synthesis of aminofurane ring; *c*) transamination reactions. In our previous paper[1] we prepared aminofurane derivatives by reduction. Stability of these derivatives necessitates the presence of a deactivating substituent in the furane nucleus. 2-Amino-3-cyano-4,5-disubstituted furanes also belong to the type of considerably stable aminofuranes[2]. These derivatives are prepared most often from acyloins[3], α-halogen ketones[4,5] or by other methods, *e.g.*[6-8].

The present work deals with synthesis of 2-amino-3-cyano-4,5-disubstituted aminofurane derivatives *IV*–*VI* by the Gewald's method from acyloins and propanedinitrile with catalysis of diethylamine; yields 55–75% (Scheme 1).

IR spectra of these derivatives showed absorption bands of amino group within 3452–3360 cm^{-1}, 3332–3320 cm^{-1}, 1667–1685 cm^{-1}, and that of CN group within 2221–2225 cm^{-1}. UV spectra of the derivatives *IV*–*VI* showed 4 absorption maxima in the regions 201–204 nm, 230–233 nm, 260–271 nm, and 320–340 nm. The λ_{max} values of the last absorption band indicate conjugation between NH_2 and CN groups through furane ring, which supports the hypothesis about stability of these derivatives[9]. Extent of the conjugation is well reflected in chemical shifts of amino group in ^{1}H-NMR spectra. The values $\delta(NH_2)$ = 7·62–7·73 ppm, compared with the earlier discussed[1,10] aminofuranes with free amino group, indicate considerable delocalization of *p* electrons of amino group.

Reaction with formamide was used for synthesis of the corresponding 4-amino-5,6-disubstituted furo[2,3-*b*]pyrimidines *VII*–*IX* with the yields 67–85% (Scheme 1).

In literature synthesis of these derivatives is mentioned in connection with a search for biologically active compounds[11-16]. The derivatives *VII–IX* showed in their IR spectra (besides others) characteristical absorption bands of NH_2 group in the regions 3465–3515 cm^{-1}, 3300–3405 cm^{-1} and 1631–1655 cm^{-1}. UV spectra showed 4 absorption maxima within 207–208 nm, 226–243 nm, 269–302 nm and 313–335 nm. The derivatives *VII–IX* are well characterized by the ^{1}H-NMR spectral data. Besides the furane[17], thiophene[17,18] and phenyl proton signals whose chemical shifts and multiplicity are analogous to those of the aminofuranes *IV–VI*, the chemical shifts $\delta(NH_2) = 6{\cdot}83-6{\cdot}00$ ppm and that of the proton of pyrimidine nucleus $\delta(H_A) = 8{\cdot}23-8{\cdot}20$ ppm stand in good accordance with literature data[18].



Comparison of the chemical shifts $\delta(NH_2)$ with those of free amino group in other systems indicates smaller extent of conjugation participation of the NH_2 group. A practically constant value of chemical shift of the pyrimidine proton H_A indicates slight influence of 5,6-substituents on the 4-aminofuropyrimidine skeleton.

Character of NH_2 group in the derivatives *IV* and *VI* is also illustrated by their reactivity to acetanhydride. The former and the latter substances gave the monoacylated (*X*) and diacylated (*XI*) aminofurane derivatives, respectively (Scheme 1). For further synthetic use the derivatives *IV–VI* were converted into the respective azomethines *XII–XIV* by reaction with aldehydes (Scheme 1, Table I). The reactivity

TABLE I

Properties of Azomethines of 2-Amino-3-cyano-4,5-diarylfuranes

No R′	Formula (mol. mass)	M.p., °C yield, %	Calculated/Found % C	 % H	 % N	ν(—C=N—) cm^{-1} [a]	λ_{max}, nm (log ε)[b]
XIIa 2-Furyl	$C_{18}H_{10}N_2O_4$ (318·3)	164—165 38	67·92 67·75	3·16 3·10	8·80 8·71	1 605	428 (4·23)
XIIb 2-Thienyl	$C_{18}H_{10}N_2O_3S$ (334·3)	110—112 41	64·65 64·37	3·01 2·99	8·38 8·40	1 590	435 (4·07)
XIIc 3-Chromonyl	$C_{23}H_{12}N_2O_5$ (396·4)	230 (dec.) 56	69·69 59·39	3·05 3·00	7·06 7·12	1 626	420 (4·10)
XIId Salicylyl	$C_{20}H_{12}N_2O_4$ (344·3)	170 48	69·76 69·71	3·51 3·48	8·13 8·10	1 600	431 (4·46)
XIIe 4-Nitrophenyl	$C_{20}H_{11}N_3O_5$ (373·3)	202 43	64·34 64·31	2·97 2·95	11·25 11·20	1 611	455 (3·62)
XIIf Piperonyl	$C_{21}H_{12}N_2O_5$ (372·3)	138—142 54	67·73 67·54	3·24 3·20	7·52 7·48	1 612	418 [c]
XIIg 5-Nitro-2-thienyl	$C_{18}H_9N_3O_5S$ (379·3)	56—60 37	56·98 57·18	2·32 2·25	11·07 11·10	1 635	495 (3·44)
XIIIa 2-Thienyl	$C_{18}H_{10}N_2OS_3$ (366·5)	158—160 72	58·98 58·90	2·75 2·70	7·64 7·71	1 589	423 (4·12)
XIIIb 3-Chromonyl	$C_{23}H_{12}N_2O_3S_3$ (428·5)	247 88	64·46 64·38	2·82 2·81	6·53 6·50	1 620	418 (4·07)
XIVa 2-Furyl	$C_{22}H_{14}N_2O_2$ (338·4)	175—180 92	78·08 78·00	4·17 4·12	8·28 8·25	1 608	422 (3·60)
XIVb 5-Nitro-2-furyl	$C_{22}H_{13}N_3O_4$ (383·3)	234—237 85	68·92 68·79	3·41 3·40	10·96 10·90	1 607	455 (4·13)
XIVc 2-Thienyl	$C_{22}H_{14}N_2OS$ (354·4)	169 90	74·55 74·50	3·98 3·91	7·90 7·93	1 595	431 (4·34)
XIVd 5-Nitro-2-thienyl	$C_{22}H_{13}N_3O_3S$ (399·4)	227—230 81	66·15 66·00	3·27 3·21	10·52 10·35	1 600	448 (3·83)
XIVe 4-Nitrophenyl	$C_{24}H_{15}N_3O_3$ (393·4)	187—189 79	73·27 73·20	3·84 3·78	10·68 10·55	1 608	430 (4·03)

TABLE I
(*Continued*)

No R'	Formula (mol. mass)	M.p., °C yield, %	Calculated/Found % C	% H	% N	ν(—C=N—) cm^{-1} [a]	λ_{max}, nm (log ε)[b]
XIVf 4-Acetaminophenyl	$C_{26}H_{19}N_3O_2$ (405·4)	244 93	77·01 77·00	4·72 4·70	10·36 10·31	1 595	426 (4·42)
XIVg Phenyl	$C_{24}H_{16}N_2O$ (348·4)	203 94	82·73 82·65	4·63 4·58	8·04 8·13	1 605	437 (3·47)
XIVh 4-N,N-Dimethyl-aminophenyl	$C_{26}H_{21}N_3O$ (391·5)	181 90	79·77 79·70	5·40 5·32	10·73 10·59	1 590	446 (4·69)
XIVi Salicylyl	$C_{24}H_{16}N_2O_2$ (364·4)	190 95	79·11 79·03	4·42 4·40	7·68 7·58	1 600	458 (4·00)
XIVj Piperonyl	$C_{25}H_{16}N_2O_3$ (392·4)	220 93	76·51 76·44	4·11 4·10	7·14 7·14	1 605	421 (4·38)
XIVk 3-Chromonyl	$C_{27}H_{16}N_2O_3$ (416·4)	232 53	77·87 77·82	3·87 3·85	6·72 6·80	1 620	411 (4·34)
XIVl 5-(3,4-Dichloro-phenyl)-2-furyl	$C_{28}H_{16}Cl_2N_2O_2$ (483·3)	186 51	69·57 69·53	3·33 3·32	5·79 5·70	1 607	440 [c]

[a] The spectra were measured in KBr disc; [b] the spectra were measured in methanol; [c] saturated solution.

decreases from aminofurane *VI* to the least reactive derivative *IV*. Similarly, stability of the azomethine derivatives decreased from the stable ones *XIV* to unstable azomethines *XII*. Analogous azomethine derivatives are synthetized[19] because of their possible biological activity.

In conclusion it can be stated that relatively high reactivity of NH_2 group contrasts with inertness of the CN group in these *o*-aminonitrile systems[20]. This system does not seem to react until under more vigorous conditions[2] or with more reactive reagents[21,22].

Preliminary tests of antibacterial and mutagenic activity of some of the above derivatives showed their inefficiency[23].

EXPERIMENTAL

The melting points were not corrected. The IR, UV, ^{1}H-NMR and mass spectra were measured with a Zeiss UR-20, Specord UV VIS, Tesla BS 487 B (80 MHz) and AEI MS 902 S, respectively.

2-Amino-3-cyano-4,5-di(furyl)furane (*IV*)

8·7 g (0·045 mol) furoin *I* and 3·85 g (0·058 mol) propanedinitrile were dissolved in 13·6 ml dimethylformamide. The mixture was stirred and 1·8 ml diethylamine was added during 30 min whereupon stirring was continued at 20°C 24 h. The mixture was poured in water, and the raw product was recrystallized from ethanol. M.p. 187—188°C, yield 61%. For $C_{13}H_8N_2O_3$ (240·2) calculated: 65·00% C, 3·36% H, 11·66% N; found; 64·46% C, 3·35% H, 11·48% N. IR spectrum (KBr), ν_{max}, cm^{-1}: 3450, 3332, 1685 (—NH_2), 2225 (CN), 1027 (C—O—C). UV spectrum (methanol), λ_{max}, nm (log ε): 201 (4·17), 230 (4·17), 260 (4·24), 333 (4·22). ^{1}H-NMR spectrum (hexadeuteriodimethyl sulphoxide), δ, ppm: 7·62 s (NH_2, 2 H), 7·71 dd, 6·55 dd, 6·78 dd and 7·66 dd, 6·53 dd, 6·67 dd (furane protons, 6 H). Mass spectrum, *m/e* (ref. intensity, %): 241 (15), 240 (100) M$^{+\cdot}$, 211 (29), 197 (12), 196 (10), 184 (13), 156 (10), 155 (21), 140 (12), 129 (8), 114 (9), 113 (8), 95 (10), 88 (9), 63 (11), 44 (12), 39 (24).

2-Amino-3-cyano-4,5-di(2-thienyl)furane[24] (*V*)

Solution of 7·4 g (0·033 mol) thienoin (*II*) and 2·8 g (0·042 mol) propanedinitrile in 10 ml dimethylformamide was stirred and treated with 1·3 ml diethylamine and left to stand 24 h. Then the solution was poured on ice, the raw product was collected by suction and purified by crystallization from ethanol; m.p. 184°C, yield 4·95 g (55·1%). For $C_{13}H_8N_2OS_2$ (272·3) calculated: 57·32% C, 2·96% H, 10·28% N; found: 57·21% C, 2·89% H, 10·13% N. IR spectrum (KBr) ν_{max}, cm^{-1}: 3452, 3320, 1667 (—NH_2), 2221 (CN), 1048 (C—O—C). UV spectrum (methanol), λ_{max}, nm (log ε): 208 (4·15), 233 (4·13), 262 (4·15), 340 (4·09). ^{1}H-NMR spectrum (hexadeuteriodimethyl sulphoxide), δ, ppm: 7·73 s (NH_2, 2 H), 7·60 dd, 7·41 dd, 6·96 dd, 7·26—7·01 a complex multiplet (thiophene protons, 6 H). Mass spectrum *m/e*, (rel. intensity, %): 274 (12), 273 (20), 272 (100) M$^{+\cdot}$, 243 (19), 230 (9), 229 (32), 228 (36), 211 (11), 202 (9), 201 (8), 196 (9), 158 (8), 145 (9), 111 (18), 110 (10), 83 (7), 69 (16), 45 (27), 44 (57), 41 (9), 39 (55).

2-Amino-3-cyano-4,5-diphenylfurane[3] (*VI*)

Solution of 8·5 g (0·13 mol) propanedinitrile and 21 g (0·10 mol) benzoin (*III*) in 30 ml dimethylformamide was treated with 4 ml diethylamine. After 12 h standing and 12 h stirring the mixture was poured into 60 ml water, the separated solid was collected by suction and recrystallized from ethanol. Yield 25 g (75%), m.p. 206°C. For $C_{17}H_{12}N_2O$ (260·3) calculated: 78·43% C, 4·64% H, 10·76% N; found: 78·30% C, 4·55% H, 10·70% N. IR spectrum (KBr) ν_{max}, cm^{-1}: 3360, 3320, 1667 (—NH_2), 2221 (CN), 1030 (C—O—C). UV spectrum (methanol), λ_{max}, nm (log ε): 204 (4·60), 231 (4·20), 271 (4·01), 320 (4·14). ^{1}H-NMR spectrum (hexadeuteriodimethyl sulphoxide), δ, ppm: 7·64 broad singlet (NH_2, 2 H), 7·37 s and 7·18 s (the phenyl protons, 10 H). Mass spectrum *m/e* (rel. intensity, %): 261 (20), 260 (100) M$^{+\cdot}$, 231 (8), 217 (16), 216 (20), 190 (14), 189 (17), 149 (9), 105 (26), 67 (24), 51 (14), 44 (8).

4-Amino-5,6-di(2-furyl)furo[2,3-*b*]pyrimidine (*VII*)

Mixture of 9·6 g (0·04 mol) aminofurane *IV* and 40 ml formamide was refluxed 50 min and poured in cold water. The raw product was collected by suction and recrystallized from ethanol to give

7·15 g (67%) derivative *VII*, m.p. 196—198°C. For $C_{14}H_9N_3O_3$ (267·2) calculated: 62·91% C, 3·39% H, 15·72% N; found: 62·86% C, 3·31% H, 15·35% N. IR spectrum (KBr) ν_{max}, cm^{-1}: 3515, 3405, 1631 (—NH_2), 1600 (C=C), 1029 (C—O—C). UV spectrum (methanol) λ_{max}, nm (log ε): 208 (4·18), 236 (4·01), 278 (3·90), 333 (4·22). ^{1}H-NMR spectrum (hexadeuteriodimethyl sulphoxide) δ, ppm: 8·23 s (H_A, 1 H), 6·83 broad singlet (NH_2, 2 H), 7·88 dd, 7·82 dd, 6·93 dd, 6·88 dd, 6·66 dd (furane protons, 6 H). Mass spectrum *m/e* (rel. intensity, %): 268 (18), 267 (100) $M^{+\cdot}$, 226 (9), 239 (11), 237 (11), 211 (23), 184 (9), 155 (8), 95 (10), 39 (8).

4-Amino-5,6-di(2-thienyl)furo[2,3-*b*]pyrimidine[25] (*VIII*)

Solution of 0·2 g (0·73 mmol) aminofurane *V* in 6 ml formamide was refluxed 30 min, cooled and poured in cold water. The obtained solid was collected by suction and recrystallized from excess ethanol to give 150 mg (71·4%) compound *VIII*, m.p. 217°C. For $C_{14}H_9N_3OS_2$ (299·4) calculated: 56·16% C, 3·03% H, 14·03% N; found: 56·08% C, 3·00% H, 14·01% N. IR spectrum (KBr) ν_{max}, cm^{-1}: 3465, 3402, 1655 (—NH_2), 1598, 1580 (C=C), 1047 (C—O—C). UV spectrum (methanol) λ_{max}, nm (log ε): 208 (4·34, 243 (4·00), 269 (3·91), 320 (4·27), 335 (4·36). ^{1}H-NMR spectrum (hexadeuteriodimethyl sulphoxide) δ, ppm: 8·20 s (H_A, 1 H), 6·30 s (NH_2, 2 H), 7·82 dd, 7·55 dd, 7·03 dd, 7·36—7·19 m (the thiophene protons, 6 H). Mass spectrum *m/e* (rel. intensity, %): 301 (10), 300 (24), 299 (100) $M^{+\cdot}$, 298 (29), 272 (10), 243 (7), 228 (7), 134 (8), 111 (18), 110 (6), 45 (6), 44 (14), 39 (14).

4-Amino-5,6-diphenylfuro[2,3-*b*]pyrimidine (*IX*)

This derivative was prepared according to ref.[3], yield 85%, m.p. 267°C. For $C_{18}H_{13}N_3O$ (287·3) calculated: 75·24% C, 4·56% H, 14·62% N; found: 75·17% C, 4·49% H, 14·70% N. IR spectrum (KBr) ν_{max}, cm^{-1}: 3468, 3300, 1655 (—NH_2), 1600 (C=C), 1032 (C—O—C). UV spectrum (methanol) λ_{max}, nm (log ε): 207 (4·62), 226 (4·30), 302 (4·39), 313 (4·46), 327 (4·31). ^{1}H-NMR spectrum (hexadeuteriodimethyl sulphoxide) δ, ppm: 8·22 s (H_A, 1 H), 6·00 broad singlet (NH_2, 2 H), 7·50—7·31 m (the phenyl protons, 10 H). Mass spectrum *m/e* (rel. intensity, %): 288 (20), 287 (100) $M^{+\cdot}$, 286 (72), 231 (7), 216 (6), 189 (9), 149 (8), 144 (7), 128 (8), 105 (7), 77 (22), 51 (7), 44 (12), 32 (53).

2-Acetylamino-3-cyano-4,5-di(2-furyl)furane (*X*)

Mixture of 1 g (0·041 mol) aminofurane *IV* and 10 ml acetanhydride was refluxed 3 h, evaporated, and the residue was separated chromatographically (Al_2O_3 column — Brockmann II, benzene–ethyl acetate 1 : 4). The main fraction gave 0·75 g (64%) substance *X*, m.p. 176—178°C. For $C_{15}H_{10}N_2O_4$ (282·3) calculated: 63·82% C, 3·57% H, 9·93% N; found: 64·25% C, 3·55% H, 9·71% N. IR spectrum (KBr) ν_{max}, cm^{-1}: 3190, 3128, 1610 (—NH—), 2239 (CN), 1692 (—CO—) 1015, 1028 (C—O—C). ^{1}H-NMR spectrum (hexadeuteroacetone) δ, ppm: 7·64 dd, 7·61 dd, 6·91 dd, 6·85 dd, 6·56 dd, 6·51 dd (the furane protons, 6 H), 2·15 s (CH_3, 3 H), —NH— overlapped.

2-(N,N-Diacetylamino)-3-cyano-4,5-diphenylfurane (*XI*)

Mixture of 4 g (0·015 mol) aminofurane *VI* and 40 ml acetanhydride was refluxed 4 h, and, after evaporation, the raw product was separated on a silica gel column (150/250) using ethyl acetate as eluent. Besides unreacted aminofurane *VI* we obtained 1·4 g (26·5%) substance *XI*, m.p.

156—158°C. For $C_{21}H_{16}N_2O_3$ (344·4) calculated: 73·24% C, 4·68% H, 8·14% N; found: 74·34% C, 4·70% H, 8·09% N. IR spectrum (KBr) ν_{max}, cm^{-1}: 2248 (CN), 1750, 1720 (C=O), 1020 (C—O—C). ^{1}H-NMR spectrum (hexadeuterioacetone) δ, ppm: 7·44—7·31 m (the phenyl protons, 10 H), 2·38 s (2 × CH_3, 6 H).

Azomethines of 2-Amino-3-cyano-4,5-di(2-furyl)furane *XIIa—XIIg*

Solution of 2·4 g (0·01 mol) aminofurane *IV* in 30 ml ethanol was treated with 0·01 mol respective aldehyde and the mixture was refluxed 1—6 h to produce yellow to red solutions. After evaporation the products were purified chromatographically (silica gel columns 150/250; chloroform as eluent). The coloured fractions were recrystallized (Table I).

Azomethines of 2-Amino-3-cyano-4,5-di(2-thienyl)furane *XIIIa*, *XIIIb*

Solution of 0·27 g (0·001 mol) aminofurane *V* in 15 ml ethanol was treated with 0·001 mol respective aldehyde and then refluxed 1 h. After evaporation the products were purified chromatographically (silica gel columns 150/250; chloroform as eluent). Crystallization gave orange substances (Table I).

Azomethines of 2-Amino-3-cyano-4,5-diphenylfurane *XIVa—XIVl*

Solution of 2·06 g (0·01 mol) aminofurane *VI* in 30 ml ethanol was treated with 0·01 mol respective aldehyde and the mixture was refluxed 1—10 h to give orange solutions. After evaporation the products were purified chromatographically (AlO_3 columns, Brockmann II; chloroform as eluent). The azomethines were obtained from the coloured fractions by crystallization (Table I).

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