^{1}H, ^{13}C, and ^{14}N NMR study of 3-methylfurazans with nitrogen-containing substituents at position 4

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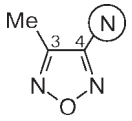
3-Methylfurazans with nitrogen-containing substituents at position 4 were studied by ^{1}H, ^{13}C, and ^{14}N NMR spectroscopy. A correlation between the chemical shifts in ^{13}C NMR spectra of these furazans and monosubstituted benzenes with the same substituents was found. The increments for a number of furazan-containing substituents were determined for the first time.

Key words: furazans, ^{1}H, ^{13}C, ^{14}N NMR spectroscopy, increments of substituents.

Furazan derivatives with nitrogen-containing substituents are valuable for practical purposes. For example, some aminofurazans exhibit a sedative effect,[1] function as antagonists for histamine H_2-receptors,[2] selective M_1-muscarine agonists,[3] and pesticides.[1,4] Azofurazans inhibit the soluble form of guanylate cyclase.[5] Some nitro- and nitraminofurazans have been proposed[6a,b] as components of explosive or pyrotechnic formulations and rocket propellants.

Structural research into the structures of new compounds by NMR spectroscopy is substantially facilitated when data on the increments of substituents for a given class of compound are available, additive schemes have been developed, and spectrum—structure correlations have been elucidated. Monosubstituted furazans,[7] phenylfurazans,[8] azoxyfurazans,[6b] and hydroxy-, alkoxy-, and phenoxyfurazans[9] have been studied previously by NMR spectroscopy. This work presents data of ^{1}H, ^{13}C, and ^{14}N NMR study for more than twenty 3-methylfurazan derivatives **1**—**22** (Table 1) with various nitrogen-containing substituents at position 4. The compounds were synthesized by previously reported procedures;[12] for compounds **16**—**18**, published data are given (see Table 1).



1—**22**

Experimental

^{1}H, ^{13}C, and ^{14}N NMR spectra were recorded on a Bruker AM-300 spectrometer (300.13, 75.47, and 21.69 MHz, respectively) at 297 K. The ^{1}H and ^{13}C NMR chemical shifts were measured relative to the solvent signals (7.27 and 77.0 ppm for $CDCl_3$; 2.05 and 30.0 ppm for acetone-d_6; 2.50 and 39.5 ppm for DMSO-d_6, respectively), and the ^{14}N NMR chemical shifts were referred to $MeNO_2$ and reported the δ scale. The ^{1}H—^{13}C spin-spin coupling constants needed for assignment of the ^{13}C NMR signals of furazans were measured by the SPT procedure.[13]

Results and Discussion

In the ^{1}H NMR spectra of the title compounds, the signal of the Me group at the furazan ring is a singlet. The signals of compounds **3** and **19**, containing two Me groups, were assigned based on examination of long-range spin-spin coupling constants between the methyl protons and the neighboring C atoms of the carbon skeleton (their assignment is considered below).

As can be seen from Table 1, the singlet of the Me group in the ^{1}H NMR spectra of compounds **1**—**22** occurs at δ 2.25—2.83. As the electron-withdrawing properties of the nitrogen-containing substituent increase, the signal shifts downfield. However, there is no clear correlation between the chemical shift and the Hammett constant of the substituent (σ_I). For example, the Me-group chemical shift for compound **12** ($\sigma_I = 0.4$) is 2.27 ppm, whereas for compound **9**, incorporating less electron-withdrawing substituent ($\sigma_I = 0.3$), this value is 2.60 ppm.

The region of the Me-group carbon in the ^{13}C NMR spectrum changes insignificantly following the variation of substituents at C(4). The chemical shifts (7.3 to 10.4 ppm) cannot be calculated satisfactorily by an additive scheme. Indeed, in the ^{13}C NMR spectra of compounds **3** and **22**, whose substituents differ crucially in electron-acceptor properties, the signals of the methyl C atom appear at 9.6 and 9.5 ppm, respectively. For compounds **3** and **19**, the ^{13}C NMR signals of the two Me groups were identified by heteronuclear double resonance with the

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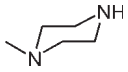
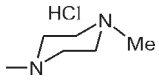
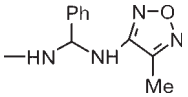
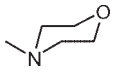
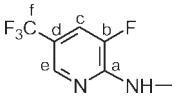
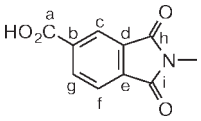
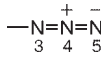
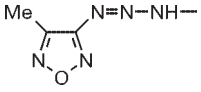
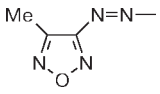
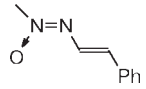
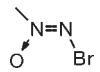
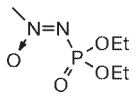
Table 1. ^{1}H, ^{13}C, and ^{14}N NMR spectra (in acetone-d_6) of 3-methylfurazans **1**—**22** wth nitrogen-containing substituents at C(4)

Compound	(N)	^{1}H NMR, δ (*J*/Hz)		^{13}C NMR, δ ($J_{C,H}$/Hz)				^{14}N NMR, δ ($\Delta\nu_{1/2}$/Hz)
		Me	The protons in (N)	Me	C(3)	C(4)	The C atoms in (N)	
1	NH_2	2.25	5.44 (br.s, 2 H)	7.7	145.4 ($^{2}J_{C,H}$ = 7.0)	157.2 ($^{3}J_{C,H}$ = 2.5)	—	—
2 [a]		2.35	2.83 (br.s, 4 H, 2 C(4)NCH_2); 3.15 (br.s, 4 H, 2 CH_2NH)	9.9	145.3	159.8	44.8 (C(4)NCH_2); 49.1 (CH_2NH)	—
3 [a]		2.35	2.75 (s, 3 H, Me); 3.20—3.50 (m, 8 H, 4 CH_2)	9.6	145.4	158.7	45.5, 51.6 (CH_2); 42.7 (Me)	—
4 [a]		2.30	6.05 (t, 1 H, CH, *J* = 6.6); 7.31—7.65 (m, 7 H, Ph + 2 NH)	9.0	144.7	155.6	69.2 (CH); 127.4, 129.6, 129.7 (C_{Ph}); 139.4 (C_{ipso})	—
5 [b]		2.35	3.25 (t, 4 H, 2 CH_2N, *J* = 5.9); 3.82 (t, 4 H, 2 CH_2O, *J* = 5.9)	10.0	144.5	159.5	48.8 (CH_2N); 66.0 (CH_2O)	—
6		2.25	8.12 (d, 1 H, H_c, *J* = 8); 8.38 (s, 1 H, H_e); 10.32 (br.s, 1 H, NH)	8.6	149.4 ($^{2}J_{C,H}$ = 7.5)	151.8 ($^{3}J_{C,H}$ = 2.5)	146.9 (C_a, $^{2}J_{C,F}$ = 18); 146.3 (C_b, $^{1}J_{C,F}$ = 384); 120.3 (C_c, $^{2}J_{C,F}$ = 24); 118.1 (C_d, $^{2}J_{C,F}$ = 49); 140.4 (C_e, $^{3}J_{C,F}$ = 7); 123.1 (C_f, $^{1}J_{C,F}$ = 437)	—
7 [a]	—NHCOPh	2.34	7.52—7.71 (m, 3 H, H_m, H_p, Ph); 8.05 (d, 2 H, H_o, *J* = 7.2); 11.33 (br.s, 1 H, NH)	8.8	149.4	151.9	128.3 (C_o); 128.7 (C_m); 132.0 (C_{ipso}); 132.8 (C_p); 166.0 (CO)	—
8 [a]	—NHCOCH_2Ph	2.25	3.78 (s, 2 H, CH_2); 7.20—7.45 (m, 5 H, Ph); 10.96 (br.s, 1 H, NH)	8.4	148.3	150.8	41.8 (CH_2); 126.7 (C_p); 128.1 (C_m); 129.2 (C_o); 134.8 (C_{ipso}); 169.9 (CO)	—
9		2.62	6.41 (br.s, 2 H, H_b); 7.32 (br.s, 2 H, H_a)	9.7	146.7 ($^{2}J_{C,H}$ = 7.1)	153.3 ($^{3}J_{C,H}$ = 3.0)	121.3 (C_a); 113.0 (C_b)	—

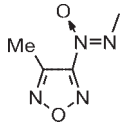
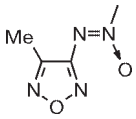
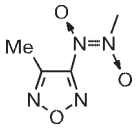
(*to be continued*)

Table 1 (*continued*)

Compound	(N)	^{1}H NMR, δ (*J*/Hz)		^{13}C NMR, δ ($J_{C,H}$/Hz)				^{14}N NMR, δ ($\Delta\nu_{1/2}$/Hz)
		Me	The protons in (N)	Me	C(3)	C(4)	The C atoms in (N)	
10 [a]		2.43	8.15 (d, H_f, J = 7.4); 8.38 (br.s, 1 H, H_c); 8.46 (br.d, H_g, J = 7.4)	8.3	150.3 ($^2J_{C,H}$ = 7.2)	146.2 ($^3J_{C,H}$ = 2.8)	124.2 (C_c); 124.7 (C_f); 132.1 (C_d); 134.6 (C_e); 136.1 (C_g); 137.1 (C_b); 164.0 (C_h, C_i); 165.7 (C_a)	—
11 [b]	—N(SO_2Me)$_2$	2.41	3.54 (s, 3 H, SO_2Me)	7.5	150.9 ($^2J_{C,H}$ = 7.3)	149.2 ($^3J_{C,H}$ = 3.2)	46.2 (SO_2Me)	—
12		2.27	—	7.3	147.2 ($^2J_{C,H}$ = 7.3)	154.4 ($^3J_{C,H}$ = 2.9)	—	30.7, 4.4 (N_{cycl}, $\Delta\nu_{1/2}$ = 550); −144.0 (N(4), $\Delta\nu_{1/2}$ = 20); −146 (N(5), $\Delta\nu_{1/2}$ = 100); −305.0 (N(3), $\Delta\nu_{1/2}$ = 700)
13		2.53	3.60 (br.s, 1 H, NH)	9.9	146.8	157.3	—	—
14	—$NHNO_2$	2.43	11.19 (s, 1 H, NH)	8.2	148.7 ($^2J_{C,H}$ = 7.2)	149.0 ($^3J_{C,H}$ = 3.0)	—	−41.0 ($NH\underline{N}O_2$, $\Delta\nu_{1/2}$ = 20)
15		2.63	—	10.3	148.6 ($^2J_{C,H}$ = 7.4)	164.0 ($^3J_{C,H}$ = 2.3)	—	—
16 [a,c]		2.69	7.40 (m, 3 H, H_m, H_p); 7.49 (d, 1 H, $\underline{H}$CPh); 7.61 (dd, 2 H, H_o); 8.59 (d, 1 H, HCN=N)	10.0	147.2	158.1	131.1 (CHN=N); 140.5 ($\underline{C}$HPh); 128.5 (C_o); 129.2, 130.8 (C_m, C_p); 134.5 (C_{ipso})	−84.0 (N→O, $\Delta\nu_{1/2}$ = 130)
17 [b,c]		2.66	—	9.3	147.1	156.1	—	−57.5 (N→O, $\Delta\nu_{1/2}$ = 20)
18 [d]		2.68	1.37 (t, 6 H, 2 Me); 4.34 (m, 4 H, 2 CH_2)	10.1	149.6	161.3	16.6 (Me); 65.8 (CH_2)	−56.0 (N→O, $\Delta\nu_{1/2}$ = 38)

(*to be continued*)

Table 1 (*continued*)

Compound	N	^{1}H NMR, δ (*J*/Hz)		^{13}C NMR, δ ($J_{C,H}$/Hz)				^{14}N NMR, δ ($\Delta\nu_{1/2}$/Hz)
		Me	The protons in N	Me	C(3)	C(4)	The C atoms in N	
19a [e]		2.83	2.56 (s, 3 H, Me)	8.6	151.5 ($^2J_{C,H}$ = 7.3)	155.7 ($^3J_{C,H}$ = 2.7)	149.3, 160.9	36.6 (N_{cycl}, $\Delta\nu_{1/2}$ = 1400); −66.5 (N→O, $\Delta\nu_{1/2}$ = 33)
19b [e]		2.56	2.83 (s, 3 H, Me)	10.4	149.3 ($^2J_{C,H}$ = 7.5)	160.9	151.5, 155.7	36.6 (N_{cycl}, $\Delta\nu_{1/2}$ = 1400); −66.5 (N→O, $\Delta\nu_{1/2}$ = 33)
20		2.58	—	7.7	147.9	152.0	—	−68.2 (N→O, $\Delta\nu_{1/2}$ = 60)
21	NO	2.65	—	7.9	137.6	171.2	—	515.1 (N=O, $\Delta\nu_{1/2}$ = 500)
22	NO_2	2.71	—	9.5	149.8 ($^2J_{C,H}$ = 7.5)	161.9 ($^3J_{C,H}$ = 2.8)	—	36.6 (N_{cycl}); −32.4 (NO_2, $\Delta\nu_{1/2}$ = 9)

[a] NMR spectra were recorded in DMSO-d_6.
[b] NMR spectra were recorded in $CDCl_3$.
[c] See Ref. 10
[d] See Ref. 11
[e] 3,3-Dimethylazoxyfurazan (**19**) containing two nonequivalent methylfurazanyl fragments is twice encountered in the Table (**19a** and **19b**).

protons of these groups (after signal assignment in the ^{1}H NMR spectra).

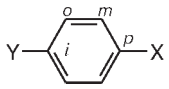
The signals of the C atoms of the furazan ring were assigned by measuring the spin-spin coupling constants of the Me-group protons with the heterocycle C atoms. The assignment was based[14] on the great difference between the spin-spin coupling constant with the C atom adjacent to the Me group ($^2J_{C,H}$ = 7.0—7.5 Hz) and the remote C atom ($^3J_{C,H}$ = 2.5—3.2 Hz). The nonequivalent furazan rings in dimethylazoxyfurazan **19** were distinguished based on the broadening of the signal for the C(4) atom proximate to the N-oxide nitrogen of one furazan ring, caused by ^{13}C—^{14}N coupling.

The signal of the C(4) atom attached to the nitrogen-containing substituent occurs at lower field (146—164 ppm) than the signal of C(3) at the Me group (145.4—151.5 ppm). Compounds **10** and **11** are exceptions; the signal of C(4) in their spectra appears at higher field than the C(3) signal. This shift can be explained by either the shielding effect of the electron clouds of the C=O and S=O groups or by the steric effect of these bulky substituents. The difference between the carbon chemical shifts of the furazan-ring atoms δ(C(4)) – δ(C(3)) amounts to 0.3—15.4 ppm and does not correlate with the electronic parameters of substituents either.

It is worth noting that the chemical shifts of the ring C atoms of 3-methyl-4-nitrosofurazan **21** are beyond the typical range. The chemical shift of the C(3) atom at the Me substituent proves to be even smaller than that of the protonated carbon in unsubstituted furazan (δ 142 ppm, *cf.* Ref. 7), whereas the chemical shift of the C atom attached to the NO group is shifted downfield by 10 ppm with respect to that of the C atom bonded to the electron-withdrawing NO_2 group. A similar influence of the NO group on the chemical shift of the C atom was reported[15] for *p*-nitrosobenzenes and interpreted by the strong π-acceptor influence of this group. Note also that in this case, the difference between the chemical shifts of the furazan C atoms (δ(C(4)) – δ(C(3)) is exceptionally great (33.6 ppm).

Previously,[7] we showed that the change in the chemical shifts of the C atoms of the heterocycle in monosub-

stituted furazans can be predicted rather accurately on the basis of an additive scheme using increments of the same substituents determined for monosubstituted benzenes. It was found that the substituent effect in the furazan ring is somewhat weaker than that in the benzene series. However, when the molecule contains several substituents, the use of simple additive schemes is known to entail, as a rule, substantial errors. Meanwhile, Lynch proposed[16] a simple linear equation (1), which relates the ^1H and ^{13}C chemical shifts in *para*-disubstituted benzenes with an invariable substituent Y and variable X to the increments of these substituents in monosubstituted benzenes:



$$Shift_X(Y) = a + b \cdot SCS_X(H), \quad (1)$$

where $Shift_X(Y)$ is the chemical shift of the C_i atom in the series of disubstituted benzenes with fixed substituent Y; $SCS_X(H)$ is the corresponding increment of the substituent X in the monosubstituted benzene (Y = H); a and b are linear regression coefficients.

This approach also proved to be efficient for *meta*- and *ortho*-disubstituted benzenes. However, it is applicable only in those cases where the steric effects of the closely spaced substituents can be neglected (see, for example, Ref. 17).

The Lynch equation was found to be useful for predicting chemical shifts of the C atoms in the 3-methylfurazans. The dependence of the chemical shifts of the C(4) atoms attached to the nitrogen-containing substituent in the furazan ring on the increments of the corresponding substituents (SCS(R)) in the *ipso*-position in monosubstituted benzenes is described by the linear equation

$$\delta(C(4))(R) = a + b \cdot SCS(R), \quad (2)$$

a = 144.9±1.4, b = 0.72±0.07,
the correlation coefficient is 0.966.

The calculation was carried out using[18] published SCS(R) values and the values that we determined from NMR-spectroscopic data for the corresponding monosubstituted benzenes.[10,11]

It can be seen from Table 2 and Fig. 1, *a* that the chemical shifts calculated using Eq. (2) are close to those found experimentally, indicating that this method can be used for the assignment of signals in the spectra of disubstituted furazans.

The coefficients appearing in Eq. (2) were determined using the chemical shifts of only 10 furazan derivatives out of the 22 compounds studied here. For other compounds, no data on the increments of the corresponding substituents in monosubstituted benzenes can be found in the literature. Using the regularity we identified, one can calculate the previously unknown SCS(R) increments for furazan-containing substituents on the basis of the chemical shifts of their C(4) atoms (Table 3).

Table 2. Correlation of the calculated and experimental chemical shifts (δ) of the C(4) atom

Compound	SCS(R), ppm	δ(C(4))		Error, ppm
		Experiment	Calculated*	
1	20.0	157.2	159.3	2.1
5	21.0	159.5	160.0	0.5
8	10.5	150.8	152.5	1.7
9	11.9	153.3	153.5	0.2
12	12.0	154.4	153.6	−0.8
16	18.2	158.1	158.0	−0.1
17	16.6	156.1	156.9	0.8
18	20.2	161.3	159.5	−1.8
21	37.6	171.2	172.0	0.8
22	20.0	161.9	159.3	−2.6

* From Eq. (2).

The use of Eq. (2) with a = 151.0 and b = 0.43 for predicting C(3) chemical shifts in furazan from known increments for the *ortho*-, *meta*-, and *para*-positions of monosubstituted benzenes did not provide satisfactory results. Even when the abnormal value for 3-methyl-4-nitrosofurazan (**21**), noted above, was neglected, the correlation coefficient between δ(C(3)) and SCS(R) did not exceed 0.84 (Fig. 1, *b*). The range of variation of the chemical shifts of C(3) in methylfurazans is relatively narrow and

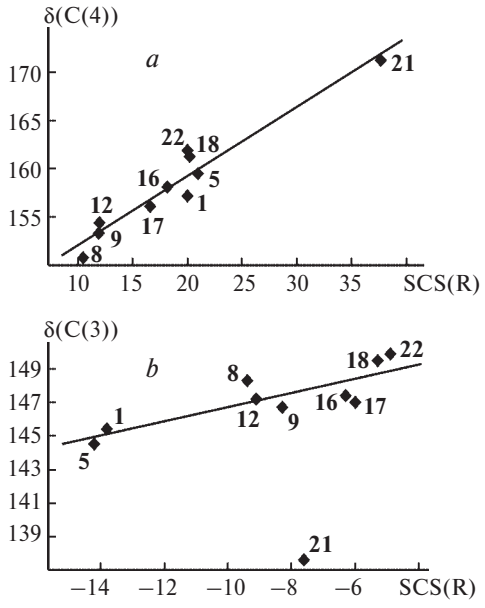
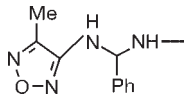
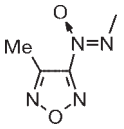
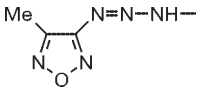
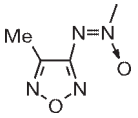
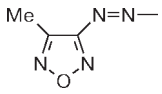
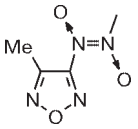


Fig. 1. Chemical shifts (δ) of the C(4) (*a*) and C(3) atoms (*b*) in substituted 3-methylfurazans *vs.* the increments of the corresponding substituents R (SCS(R)) in monosubstiuted benzenes. The straight line corresponds to Eq. (2) (a = 144.9, b = 0.72 (*a*) and a = 151.0, b = 0.43 (*b*)).

Table 3. Calculated values of the SCS(R) increments of furazan-containing substituents

Compound	Substituent	Increment SCS(R), ppm	Compound	Substituent	Increment SCS(R), ppm
4		14.9	**19***		14.9
13		17.2	**19***		22.2
15		26.5	**20**		9.9

* See note "*e*" to Table 1.

Table 4. Chemical shifts (δ) in the ^{1}H and ^{13}C NMR spectra of **1** and **22** in different solvents

Solvent	δ_H	δ_C			Δδ	
	Me	Me	C(3)	C(4)	δ(C(3)) − δ(Me)	δ(C(4)) − δ(C(3))
				1		
$CDCl_3$	2.22	7.4	144.0	155.3	136.6	11.3
Acetone-d_6	2.25	7.7	145.4	157.2	137.7	11.8
Py-d_5	2.08	7.8	145.6	157.5	137.8	11.9
DMSO-d_6	2.18	7.4	144.8	156.5	137.4	11.7
				22		
$CDCl_3$	2.67	8.9	147.5	159.7	138.6	12.2
Acetone-d_6	2.71	9.5	149.8	161.9	140.3	12.2
Py-d_5	2.42	9.1	148.9	161.2	139.8	12.3
DMSO-d_6	2.62	9.1	149.0	160.9	139.9	11.9

depends little on the nature of the substituent at the neighboring C atom, which points to an invariable pattern of electron density distribution in the furazan ring. A similar conclusion was drawn in a study of the effect of the nature of substituents on the acid—base properties of furazans.[19]

The shift of NMR signal induced by replacement of solvents is often more pronounced than that observed on passing from one substituent to another. Therefore, the solvent effect was studied separately in relation to 3-amino-4-methylfurazan (**1**) and 3-methyl-4-nitrofurazan (**22**), containing an electron-donating and an electron-withdrawing substituent, respectively. In both cases, we did not identify any specific solvation of separate groups in the molecule (Table 4). The upfield or downfield shifts of the whole spectra can apparently be explained by the inconsistency of the relative scales of chemical shifts in different solvents.

For compounds containing a polarized N atom (azoxy and nitro derivatives), Table 1 also presents ^{14}N NMR data. The signal of the N atom of the nitro group at the furazan ring is known[6b] to be a narrow singlet (compound **22**, $\Delta\nu_{1/2}$ = 9 Hz). Note that the signal of the neighboring C atom in the ^{13}C NMR spectrum is substantially broadened due to the $^{13}C—^{14}N$ coupling. A similar pattern, although less clearly defined, is observed in the spectra of azoxyfurazans **16**—**20**.

Thus, we demonstrated that the Lynch equation is, in principle, efficient in the assignment of signals of disubstituted furazans based on the extensive data available for substituted benzenes.

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