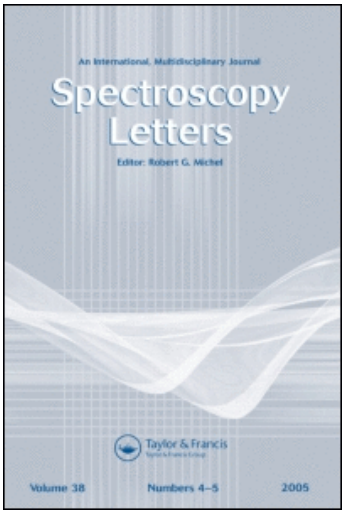

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Spectral Characteristics of the Reaction Products of 5-Phenyl-2,3,4-furantrione with o-Diamines

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SPECTRAL CHARACTERISTICS OF THE REACTION PRODUCTS OF 5-PHENYL-2,3,4-FURANTRIONE WITH o-DIAMINES.

Keywords : 6,7-Dimethylquinoxaline, quinoxaline-2-one, hydrazone, furantrione, ^{1}H and ^{13}C NMR spectra, mass spectra.

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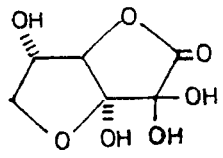
ABSTRACT

The ^{1}H and ^{13}C NMR and mass spectra of 2-(2-amino-4,5-dimethylphenylcarbamoyl)-3-(hydroxyphenylmethyl)-6,7-dimethylquinoxaline (2), 3-(hydroxyphenylmethyl)-6,7-dimethylquinoxalin-2-carboxylic-γ-lactone (5), 3-(hydroxyphenylmethyl)-6,7-dimethylquinoxalin-2-carboxylic acid phenylhydrazide (6), 3-[2-hydroxy-2-phenyl-1-(phenylhydrazono)ethyl]-6,7-dimethyl-2(1H)-quinoxalinone (7), 2,3-dihydro-6,7-dimethyl-3-phenylhydrazono-2-phenylfuro[2,3-b]quinoxaline (8), 3-(hydroxyphenylmethyl)-6,7-dimethyl-1-phenylflavazole (9), and 3-(acetoxyphenylmethyl)-6,7-dimethyl-1-phenylflavazole (10) have been studied.

INTRODUCTION

The reaction of dehydro-L-ascorbic acid (I) and its D-erythro analogue II with o-phenylenediamine gives a variety of products depending upon the molecular proportion of the reactants[1-12]. The structure of products were a subject of controversy. The products resulting from the condensation with a one molar equivalent of the diamine showed the phenomenon of fluorescence, and this property was used for their detection and determination[13-15]. The corresponding derivative from the D-erythro analogue was found to exist in the cyclic form as a mixture of the corresponding furanosyl anomers[10]. The reaction of 5-phenyl-2,3,4-furantrione (3) and o-phenylenediamine was studied[3]. When the ratio of the reactants was 1:2, the product **11a** was initially[1,2] adopted for the product, afterwards Hasselquist[5] proposed the structure **12a**, later on both structures were abandoned[4,5] in favor of structure **2a**. Similarly, the structures of the products from the L-threo and D-erythro analogues were determined and the structures like Schiff's base derivatives or having tricyclic rings were ruled out[10]. The mode of the reaction of the D-erythro analogue with o-diamines was deduced from its sequential reaction with two different diamines, whereby its structure played a major role on the cyclisation of the second amino group[16,17]. The main objective of the present work was to study the spectral characteristics of the products from the reaction of 3 with o-diamine.



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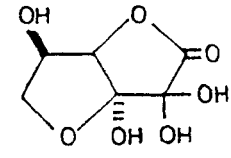

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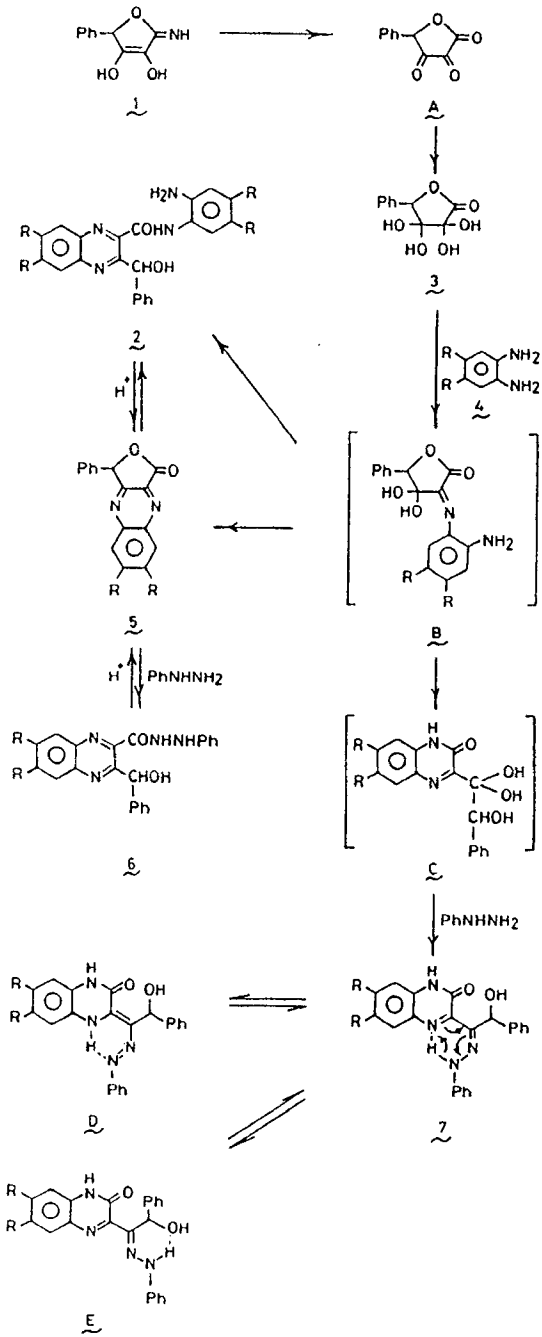
RESULTS AND DISCUSSION

Structure of 5-phenyl-2,3,4-furantrione. Examination of the structure of 5-phenyl-2,3,4-furantrione (A) by proton decoupled ^{13}C NMR spectroscopy indicated the presence of its carbonyl groups in the hydrated form as in 3. It displayed a resonance at δ_C 173.05 which was assigned to carbonyl carbon C-2, whereas the signals of C-4 and C-3 appeared at δ_C 92.27 and 96.50 indicating their hydration and ruling out their presence as carbonyl groups as in A. The resonance at δ_C 83.07 was assigned to C-5. The carbon atoms of the phenyl group showed resonances more than the expected number. This may be a consequence of the presence of 3 as a mixture of diastereoisomers at C-5.

Reaction products and pathways. A closer examination of the reaction of 3 with 4 and trapping the presumably formed products by treatment with phenylhydrazine led to the isolation of 7 in addition to 6. Their isolation could be in agreement with the intermediacy of B as proposed for the analogous pathway of the D-*erythro* analogue[16,17]. However, the subsequent cyclisation in the present case may take place with both of the two carbonyl groups at C-2 and C-4



R = Me
series a, R = H

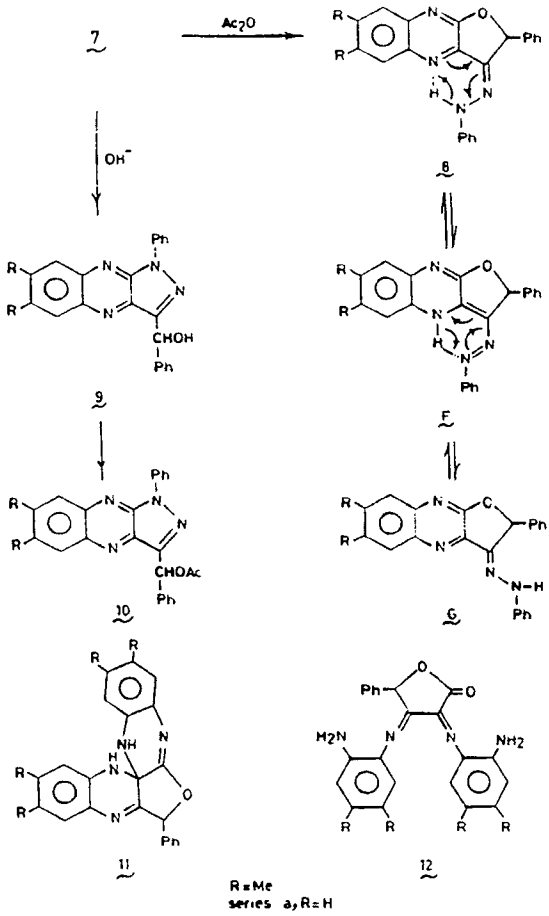
Scheme 1

to give C and 5 respectively (Scheme 1). This is in contrary to the D-erythro analogue since the latter does not give a product analogous to 6. This might be explained by the difference in reactivity of C-4 where the corresponding position of the D-erythro analogue is involved in a hemiacetal ring giving a bicyclic structure[1,2]. The competitive reactivity of C-2 and C-4 towards the cyclisation could be deduced from the percentage yields of 7 and 6 respectively, which reflects the relative formation of C and 5. This indicated the higher involvement of C-2 in the cyclisation step.

Reaction of 5 or 3 with one and two molar equivalents of 4 respectively, gave the same product 2. Its formation from 3 may be a result of the reaction of the second molecule of the diamine with either B or 5 to give 2. It should be noted that the analogous product of 5 has not been isolated from the D-erythro analogue. On the other hand, 5 could be prepared from 2 by acid hydrolysis.

The hydrazone 7 could be dehydrated in two ways. The acylating agents convert it to the furoquinoxaline 8. This could be considered as a general method for their formation. On the other hand, the action of dilute alkali on 7 gave the flavazole 9 which upon acetylation gave 10 (Scheme 2).

^{1}H NMR Spectra. The ^{1}H NMR spectra (Table 1) of the reaction products showed the presence of singlets in the range δ 2.19-2.20, and 2.42-2.61 due to the methyl groups. Closer



Scheme 2

examination of these chemical shifts led to the conclusion that these shifts could be correlated respectively with the nature of the linkage of the diamine moiety, whether it is amide, lactone, quinoxalinone or quinoxaline rings. The H-5 (δ 5.45) on the parent ring 3 appeared at the same range of the corresponding proton (δ 5.40) of 1. The product 7

Table 1.

^{1}H NMR spectra for compounds 2-10.

Compd. No.	Assignment (Chemical shifts (ppm))							
	Ac	Me	CHO (J,Hz)	Quinoxaline	2Ph	NH_2	NH (J,Hz)	OH (J,Hz)
2		2.19,2.20(2 s) 2.58,2.61(2 s)	6.70(d) (9.0)	7.92,8.01(2 s)	7.22-7.42 (s,1H),(s,1H),(m,5H)	3.33(bs)	9.50(s)	5.95(d) (9.0)
5		2.55,2.56(2 s)	6.62(s)	7.95,8.10(2 s)	7.38-7.51 (m,2H),(m,3H)			
6		2.44,2.46(2 s)	6.66(s)	7.82,7.86(2 s)	6.68-7.27 (t,2H),(t,1H),(m,4H), (m,3H)		6.21(d),9.39(s) (4.5)	5.82(d) (7.5)
7		2.47,2.48(2 s)	5.91(s)	7.55,7.63(2 s)	6.55-7.42 (s,m,2H),(m,8H)		10.83,11.14,12.22 (3 s)	5.78(d) (7.5)
8		2.42,2.43(2 s)	6.48(s)	7.63,7.79(2 s)	6.99-7.60 (m,2H),(m,2H),(d,2H) (m,2H),(m,2H)		12.10(s)	
9		2.47,2.49(2 s)	6.56(d) (6.0)	8.30,8.41(2 d) < 1	7.26-7.93 (m,2H),(t,2H),(t,2H), (d,2H),d,2H)			4.39(d) (6.0)
10	2.26(s)	2.52,2.53(2 s)	7.26(s)	8.40,8.44(2 d) < 1	7.28-8.05 (m,3H),(t,3H),(2d,2H), (2 s,2H).			

showed that proton at δ 5.91 whereas the other products 2, 6, 8 and 9 showed it at lower field in the range δ 6.23-6.70. This could be a consequence of the involvement of the hydroxyl group geminal to that proton in either hydrogen bonding with the nitrogen of the quinoxaline and flavazole rings as in 2, 6 and 9 respectively, or in a cyclic form as in 8. On the other hand, that hydroxyl group in 7 could not form such type of hydrogen bonding as a consequence of involvement of the NH of the hydrazone residue in hydrogen bonding with the nitrogen of the quinoxaline ring. Rupture of this type of hydrogen bonding in 7 or 8 by the solvent, which was used for measuring the spectra, may cause the appearance of tautomeric forms in its solution. The azo-ene structure of type D or F may be proposed as given by Kurasawa[18] for similar compounds. However, syn- and anti-structures of the type 7 ⇌ E and 8 ⇌ G may be given as reported for the corresponding methyl analogue[19].

The two protons of the quinoxaline ring are characterized by their presence at a lower field than the aromatic protons, and appeared as two singlets or two doublets with very small para coupling constants. The rest of the spectra agreed with the structures.

^{13}C NMR Spectra. The proton decoupled ^{13}C NMR spectra (Table 2 and 3) of the reaction products can be divided into two types where the diamine moiety is attached to C-2 and C-3 or C-3 and C-4 of the furantrione. The signals at a high

field in the range δ_C 19.95-20.81 were assigned to the methyl groups. Comparing those of 5 with the four signals of 2 in that region, helped in identifying the resonances of the methyl group attached to the quinoxaline ring. Close to that region is the resonance (δ_C 21.21) of the acetyl methyl of 10. The ^{13}C resonance of the carbonyl groups appeared at the lowest field region and were readily identified. The C-5 of 3 appeared at δ_C 83.07 and the respectiv carbon of 5 and 8 appeared at δ_C 81.28 and 82.83. The corresponding carbon in the other compounds belong to uncyclized derivatives of the hydroxymethine type and appeared in the range δ_C 71.52-74.51. They may be divided to two types; when that carbon attached to a quinoxaline ring, it appears at a lower magnetic field (δ_C 73.57 and 74.51) than the respective carbon (δ_C 71.52 and 71.71) attached to the flavazole ring.

Examination of the reported[20] assignment of the carbon spectrum of a model compound 6,7-dimethylquinoxaline (13) indicated that the most deshielded carbons were C-2 and C-3. A tentative assignment of carbons of the studied compounds could be done by considering the substituents on those positions in the first series 2, 5 and 6 (Table 2) and correlating them with the incremental shifts of the aromatic carbon atoms of monosubstituted benzene which can be approximated by applying the principle of substituent additivity. Thus, compound 6 has -$\underset{\text{Ph}}{\text{C}}$HOH on C-3 and -CONHNHPh on C-2. The former may cause a substantial downfield shift

Table 2.

^{13}C NMR spectra for compounds 2-6.

	Compound No.			
Assignment	13[a]	2	5	6
Me		18.79		
Me		19.40		
Me		20.40	20.50	20.32
Me		20.64	20.81	20.58
H-C-O-		74.51	81.28	73.57
C=O		162.68	166.45	164.44
Quinoxaline ring				
C-2	143.77	155.58	145.36	147.28
C-3	143.77	158.02	158.04	155.13
C-5[c]	128.16	138.58	129.72	129.80
C-6[b]	140.23	142.82	143.33	141.85
C-7[b]	140.23	142.70	136.88	140.67
C-8[c]	128.16	138.37	129.41	130.60
C-9	141.74	143.71	142.87	142.26
C-10	141.74	145.28	143.45	143.83
Phenyl group				
C'-1		136.03	134.56	138.43
C'-2,6		127.55	126.75	127.17
C'-3,5		128.44	128.96	127.78
C'-4		128.14	128.42	127.23
Phenyl group on the hydrazone or diamine residues				
C"-1		128.97		140.62
C"-2		134.58		113.44
C"-3		126.24		127.78
C"-4		129.72		121.12
C"-5		127.26		127.78
C"-6		127.22		127.44

[a] Compound 13 is 6,7-dimethylquinoxaline[20].

[b,c] Assignments may be interchanged.

of the resonance of C-3 compared to that of C-2, and a slight upfield shift may be experienced on C-2, C-9 and C-10. On the other hand, that group on C-3 may cause a substantial downfield shift of the resonance of C-10.

Compound 5 could be considered as 6 but having a fused lactone ring to C-2 and C-3 positions. The lactone ring may affect those positions as it does an ester and alkyl ester respectively in addition to the nature of being cyclic. It can be concluded that a downfield shift of the resonance of C-3 may be more than that of C-2. While that group on C-2 may not cause an effect on other carbons, that on C-3 may cause a downfield shift on C-10.

The carbon atoms of 2 were assigned by comparison with the spectrum of 6,7-dimethylquinoxaline (13) as well as by similar arguments that were used for compound 6.

The carbon assignments of the second series of compounds 8-10 were collected in (Table 3). The carbons of the phenyl rings were assigned by comparison with each other and with the parent compound. The carbons of the phenylhydrazone residues were tentatively assigned by a comparison with the respective spectra of acetone and acetophenone phenylhydrazones.

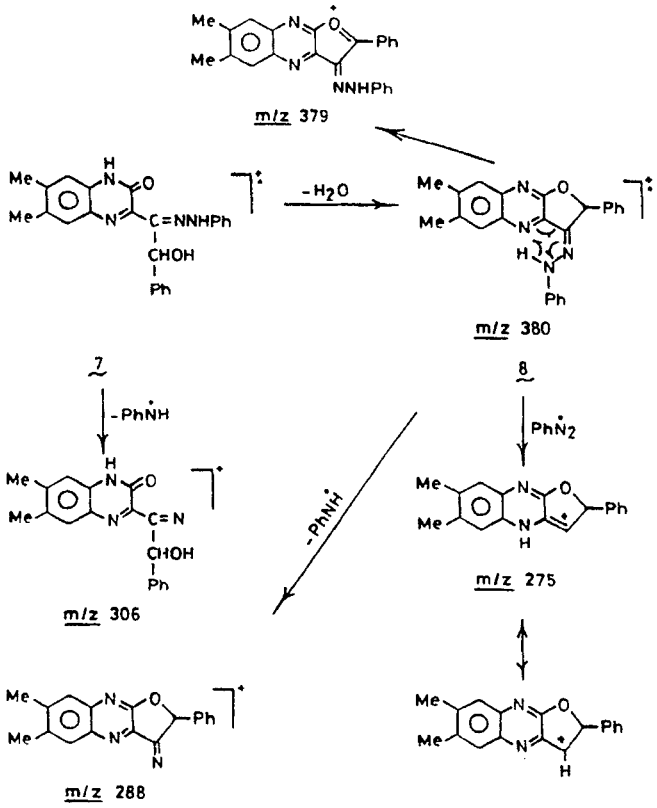
Mass Spectra. The mass spectrum of 7 did not show the molecular ion peak at m/z 398; but the highest mass ion appeared at m/z 380 due to the loss of a molecule of water (Scheme 3). The structure of that ion was given as 8 which

Table 3.

^{13}C NMR spectra for compounds 8-10.

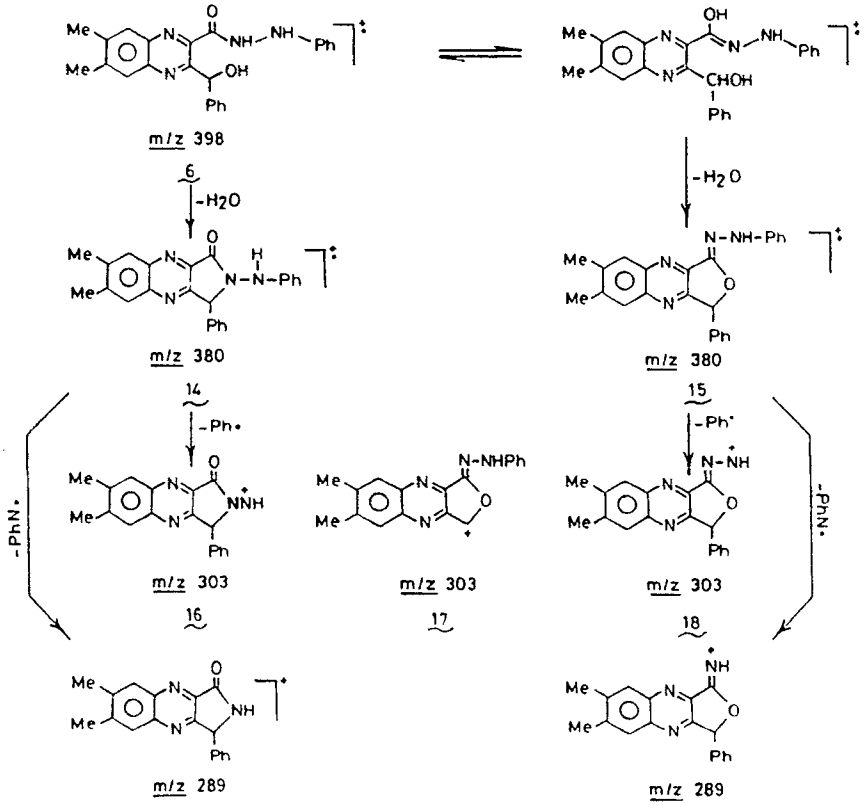
Assignment	Compound No. 8	9	10
Me	19.95	20.24	20.28
Me	20.42	20.61	20.66
Me			21.21
H-C-O-	82.83	71.52	71.71
C=O			170.24
C=N	143.38	147.30	144.46
Quinoxaline ring			
C-2	138.01	142.77	142.49
C-3	137.77	142.54	142.46
C-5c	126.46	126.83	127.66
C-6b	137.71	141.59	139.36
C-7b	137.24	139.97	139.08
C-8c	128.81	127.66	127.66
C-9	139.35	139.32	137.60
C-10	137.77	139.22	137.60
Phenyl group			
C'-1	135.11	134.91	135.06
C'-2,6	128.11	127.90	128.03
C'-3,5	129.36	129.11	129.11
C'-4	128.71	128.71	128.55
Phenyl group on the hydrazone			
C"-1	141.13	140.75	140.72
C"-2,6	113.01	119.78	120.10
C"-3,5	127.35	128.48	128.49
C"-4	122.05	125.55	125.67

[b,c] Assignments may be interchanged.



Scheme 3

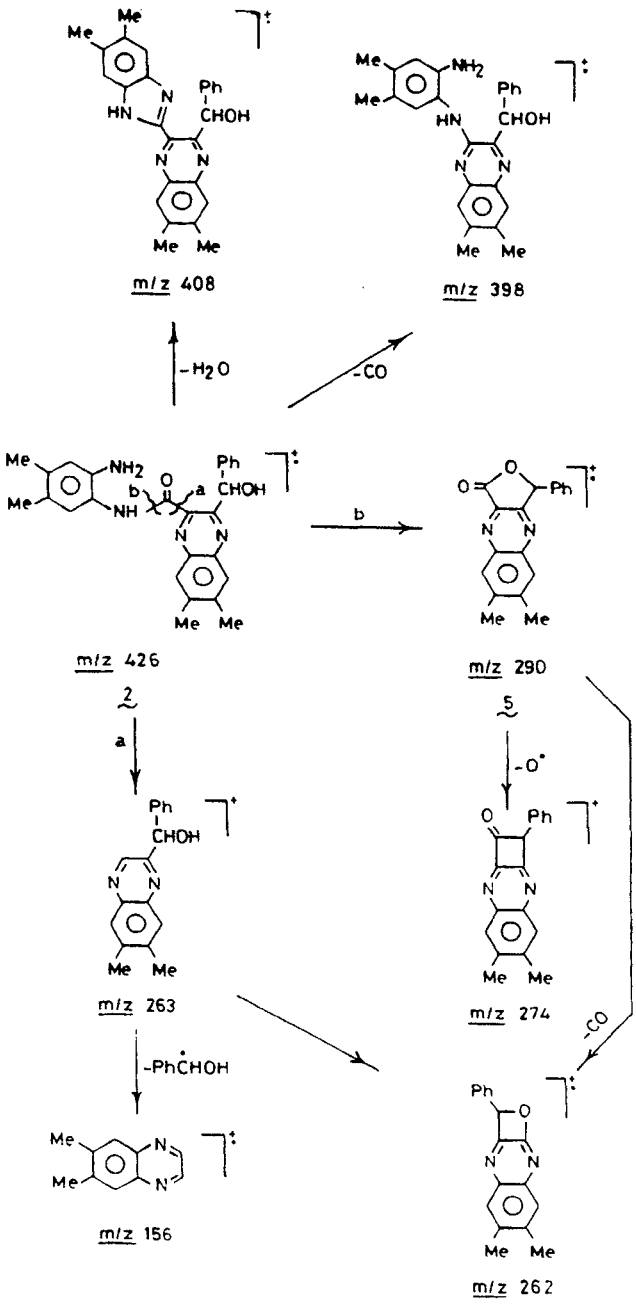
is the molecular ion peak of compound 8. Thus the rest of spectra of 7 and 8 are identical, except for an ion at m/z 306 due to the loss of PhNH from 7; the loss of that fragment from 8 gave ion at m/z 288. Transfer of a proton from the hydrazone residue to the quinoxaline ring in 8 followed by a loss of PhN_2 gave an ion at m/z 275 which was the third largest peak in the spectrum. The latter ion



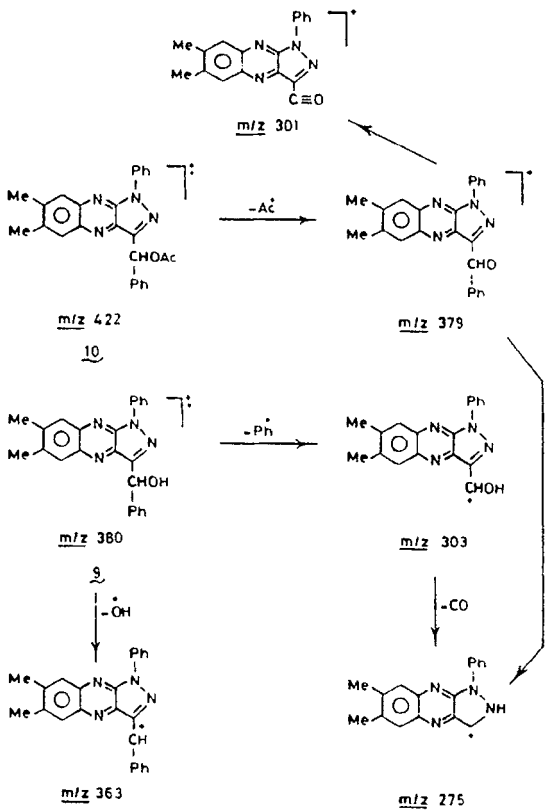
Scheme 4

underwent variety of losses to give ions at m/z 257, 246, and 169.

The hydrazide 6 did not show the molecular ion peak at m/z 398, but a loss of water molecule gave the ion at m/z 380 which could be assigned the structure 14 or 15. Further loss of Ph gave the second largest peak at m/z 303 which may have the three structures 16-18 (Scheme 4).



Scheme 5



Scheme 6

The spectra of 5 and 2 showed the highest mass ion at the same m/z 290 which it was corresponding to the molecular ion peak of 5. The spectrum of 2 showed that ion m/z 290 as a result of the loss of diamine from 2. The rest of the two spectra are almost identical except for extra ions for 2 at m/z 263 resulting from a cleavage of the diamine and a carbonyl group, whose further cleavage of the side chain gave m/z 156 (Scheme 5). Another series of

Table 4.

Mass spectra for compounds 2 - 10.

Compd. No.	m/z (Relative intensity, %)
2	426(41), 408(66), 398(27), 380(41), 364(14), 331(14), 319(40), 303(26), 302(26), 290(100), 263(55), 262(18), 261(25), 245(45), 234(40), 219(82), 204(14), 156(55), 147(96), 136(52), 128(27), 121(19), 119(41), 105(19) and 77(25).
5	290(100), 246(16), 245(19), 234(21), 157(4), 136(6), 116(1), 105(2), 89(4) and 77(25).
6	380(58), 303(89), 289(1), 275(25), 274(16), 247(14), 246(28), 231(6), 157 (5), 105(31), 93(11), 77(100) and 65(7).
7	380(86), 306(9), 275(80), 257 (1), 247(7), 231(5), 116(13), 105(47), 91(12), 77(100) and 65(19).
8	380(73), 379(27), 275(7), 257(1), 247(8), 231(5), 116(16), 105(45), 91(11), 77(100) and 65(23).
9	380(54), 363(12), 351(2), 303(8), 275(89), 273(52), 248(13), 232(1), 156(1), 116(4), 105(26), 91(5), 77(100) and 65(26).
10	422(18), 379(100), 363(28), 301(1), 275(37.65), 248(1), 180(1), 116(4), 105(12), 89(4), 77 (38), and 65(1).

ions appeared in the spectrum of 2 which resulted from the loss of H_2O from the molecular ion peak followed by loss of CO_2.

The spectra of 9 and 10 showed the respective molecular ion peaks at m/z 380 and 422. Loss of Ac and

CH_2=C=O from **10** gave ions at m/z 379 and 380 respectively. The former ion was the second largest peak for **10** whereas that ion of **9** was at m/z 275 and corresponding to the loss of the side chain at position 3. The ions at m/z 273, 274, and 275 are the ions appearing in the series of nucleoside analogues and assigned as B, B+1, and B+2 where B is the heterocyclic ring (Scheme 6). The mass spectral data of the compounds are given in Table 4.

EXPERIMENTAL

Melting points were determined with a Meltemp apparatus and are uncorrected. ^{1}H NMR spectra were determined with a Varian EM-390 and XL 300 spectrometers for solutions in chloroform-d or dimethylsulfoxide-d_6 with tetramethylsilane ($SiMe_4$) as a reference. ^{13}C NMR spectra were determined with a Varian XL 75 spectrometer. The spectra are reported with chemical shifts (ppm) downfield from ($SiMe_4$). Mass spectra were recorded with a Finnigan 4021B mass spectrometer, with an ionization energy of 70 ev, at temperature in the ion source 250. The details of a part of the experimental results will be published elsewhere[21]. Microanalyses was made in the Unit of Microanalysis, Cairo University.

2-Hydroxy-4-phenyl-tetronimide (1).

It was prepared by method of Dahn, m.p. 174-177° (lit.[3] m.p. 173-177°); ^{1}H NMR (Me_2SO-d_6) δ_H: 3.95 (bs, 2 H, 2 OH), 5.40 (s, 1 H, H-4), 7.22-7.41 (m, 5 H, Ph), and 8.70 (s, 1 H, NH).

5-Phenyl-2,3,4-furantrione (3).

To a suspension of compound 1 (1.91 g, 0.01 mol) in acetone (15 mL) and 2 N sulphuric acid (25 mL), a solution of 10% sodium nitrite (15 mL) was added drop by drop with stirring at 0°. The reaction mixture was left overnight. The product was washed with water, dried and crystallized from ethanol, m.p. 126-129° (lit.[7] m.p. 125-136°); ^{1}H NMR (Me_2SO-d_6) δ_H: 3.69 (s, 4 H, 4 OH), 5.45 (s, 1 H, H-5), and 7.55 (s, 5 H, Ph).

3-[2-Hydroxy-2-phenyl-1-(phenylhydrazone)ethyl]-6,7-dimethyl-2(1H)-quinoxalinone (7) and 3-(hydroxyphenylmethyl)-6,7-dimethylquinoxalin-2-carboxylic acid phenylhydrazide (6).

A solution of compound 3 (2.53 g, 0.01 mol) in methanol (10 mL) was treated with a solution of 1,2-diamino-4,5-dimethylbenzene (1.6 g, 0.01 mol) in methanol (5 mL), the mixture was boiled for 2 min. on a water-bath, and phenylhydrazine (1.1 g, 0.01 mol) was then added. The mixture was further boiled under reflux for 10 min, and kept overnight at room temperature. The red hydrazone 7 that separated out was filtered off, and recrystallized from ethanol (yield: 3.02 g, 80%), m.p. 235-237° (lit[21]. 235-237°). The hydrazide 6 was isolated from the mother liquor in yellow needles (yield 0.38 g, 10%). It was recrystallized from ethanol, m.p. 193-195°; IR 3380 (OH), 3280 and 3080 (NH), 1700 (CON), 1680 sh (C=N) and 1602 (C=C). Elem. anal. found % (Calcd. for $C_{24}H_{22}N_4O_2$): C, 72.6 (72.3); H, 5.6 (5.6); N, 13.8 (14.1).

3-(Hydroxyphenylmethyl)-6,7-dimethyl-1-phenylflavazole, (9)[21] was prepared from 7 by the action of 0.01 N solution of sodium hydroxide, m.p. 150-153°. **3-(Acetoxyphenylmethyl)-6,7-dimethyl-1-phenylflavazole (10)**[21] was prepared by the acetylation of 9, m.p. 136-138°. **2,3-Dihydro-6,7-dimethyl-3-phenylhydrazono-2-phenylfuro[2,3-b]quinoxaline (8)**[21] was prepared by the action of acetic anhydride in pyridine on 7, m.p. 225-227°. **2-(2-Amino-4,5-dimethylphenylcarbamoyl)-3-(hydroxyphenylmethyl)-6,7-dimethylquinoxaline (2)**[21] was prepared from 3 by reaction with two equivalents of **4b**, m.p.192-195°. **3-(Hydroxyphenylmethyl)-6,7-dimethylquinoxalin-2-carboxylic-γ-lactone (5)**[21] was prepared by refluxing a solution of 2 (0.1 g, 0.23 mol) in methanol (20 mL) and hydrochloric acid (1 mL) for 2 h. The product was found to be identical with that obtained from the reaction of 3 with a molar equivalent of **4**. Its m.p. and mixed m.p. 203-205°.

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