The benzene solubles were recovered in the usual manner to give 0.73 g. of polymer, m.p. 110–120°, mol. wt. 800. The benzene insolubles were taken up in acetonitrile and precipitated by addition of benzene to give a white solid, m.p. 140–145°. A second precipitation from acetonitrile raised the melting point to 152–154°. *Anal.* Calcd. for $[(CH_3)_2NC(H)N(CH_3)_2]Cl$: Cl, 26.0; N, 20.5. Found: Cl, 25.7; N, 20.7. The infrared spectrum of the salt in $CHCl_3$ was identical with that of a sample of tetramethylformamidinium chloride prepared according to a literature method.[19]

The experimental ratios are: $DMF/(CH_3)_2NH_2Cl = 1.03$ and $(CH_3)_2NH/(CH_3)_2NH_2Cl = 2.02$. Since two $>BN(CH_3)_2$ groups are consumed per DMF molecule, the fraction of $>BNMe_2$ groups consumed in the reaction, or the extent of reaction, p, $= 7.30/9.19 = 0.797$. The number average degree of polymerization, $\bar{P}_n = 1/1 - p$,[20] is then $1/1 - 0.797$ or 4.93. This $\bar{P}_n$ corresponds to an average molecular weight of about 750, the exact weight depending on the nature of the end groups.

Isolation and Identification of Trimethylformamidinium Chloride.—A mixture of 6.31 mmoles of B-dimethylamino-B-dimethyl-N-trimethylborazine, 3.06 mmoles of methylamine hydrochloride, and 2.71 ml. of DMF was stirred at 25° for 18 hr. After fractionation of the volatiles, 6.01 mmoles of dimethylamine, v.p. 44 mm. at −45.2°, 565 mm. at 0° (lit. 565 mm.), and 2.52 ml. of DMF were obtained. The nonvolatile residue was extracted with benzene to give 0.96 g. (96% yield) of bisborazyl oxide. The benzene insolubles were dried under vacuum to give trimethylformamidinium chloride, m.p. 129–131°. *Anal.* Calcd. for $[(CH_3)_2NC(H)NHCH_3]Cl$: Cl, 29.0; N, 22.9. Found: Cl, 29.3; N, 22.9. The infrared spectrum of the salt confirmed the amidine structure.

Reaction of B-Bis-(dimethylamino)-B-methyl-N-trimethylborazine with Aniline Hydrochloride and DMF.—The reaction of 4.72 mmoles of the borazine, 3.71 mmoles of aniline hydrochlo-

ride, and 2.62 ml. of DMF gave 6.95 mmoles of dimethylamine (v.p. = 44 mm. at −45.2°) and 2.30 ml. of excess DMF. The usual work-up gave 0.70 g. (98% yield) of poly-(tetramethylborazylene) oxide (identified through infrared spectrum) which melted over the range 110–120°. The benzene insolubles were reprecipitated from acetonitrile to give a white powder, m.p. 226–228°. *Anal.* Calcd. for $[C_6H_5NHC(H)N(CH_3)_2]Cl$: Cl, 19.2; N, 15.2. Found: Cl, 19.9; N, 16.4.

Infrared Spectra.—Spectra of the borazyl oxides were obtained on CCl_4 solutions. Since the amidine salts are insoluble in CCl_4, spectra of these compounds were obtained on chloroform solutions. The prominent absorption bands in cm.$^{-1}$ of the compounds examined are

$O[(CH_3)_2[BN]_3(CH_3)_3]_2$: 2960 m, 2930 m, 2850 w, 1450 s, 1410 s, 1383 vs, 1332 m, 1282 m, 1250 m, 1121 m, 1022 w, 946 w, 882 m, 697 w, 674 m, 663 w

$[O(CH_3)[BN]_3(CH_3)_3]_x$: 2970 m, 2930 m, 2860 w, 1454 s, 1402 vs, 1382 vs, 1350 s, 1272 m, 1253 w, 1209 m, 1191 w, 1110 m, 1053 m, 1021 w, 955 w, 883 w, 693 w

$[O(C_4H_9)[BN]_3(CH_3)_3]_x$: 2930 m, 2860 m, 1455 s, 1400 vs, 1380 vs, 1343 s, 1277 w, 1259 w, 1209 w, 1104 w, 1058 w, 1025 m, 952 w, 935 w, 699 w, 663 vw

$O[(CH_3)_2[BN]_3(C_2H_5)_3]_2$: 2980 m, 2930 m, 2870 w, 2780 vw, 1430 s, 1393 s, 1379 m, 1360 m, 1317 w, 1292 s, 1245 vw, 1212 vw, 1144 vw, 1119 m, 1091 m, 1048 n, 897 w, 884 m, 681 w

$[O(CH_3)[BN]_3(C_2H_5)_3]_x$: 2980 m, 2930 m, 2875 w, 1414 s, 1378 s, 1362 s, 1291 s, 1255 w, 1216 w, 1181 vw, 1118 w, 1091 w, 1048 m, 894 vw, 881 w, 698 w

$[(CH_3)_2NC(H)N(CH_3)_2]Cl$: 3310 m, 2880 s, 2700 w, 2430 m, 1703 s, 1490 m, 1449 s, 1401 s, 1271 m, 1230 m, 1160 m, 1108 w, 1087 w, 1052 w, 1007 vw, 880 vw, 865 m

$[(CH_3)_2NC(H)NHCH_3]Cl$: 3320 m, 3110 m, 2880 s, 2760 s, 2430 m, 2270 w, 1718 vs, 1621 vw, 1588 vw, 1488 w, 1446 s, 1368 s, 1265 w, 1140 s, 1084 w, 1063 m, 1005 m, 881 w, 858 m

(19) Z. Arnold, *Collection Czech. Chem. Commun.*, **24**, 760 (1959).

(20) H. Mark and A. V. Tobolsky, "Physical Chemistry of High Polymeric Systems," 2nd Ed., Interscience Publishers, Inc., New York, N. Y., 1950, pp. 368–369.

The Aporphine and Isoquinolinedienone Alkaloids of *Ocotea glaziovii*[1]

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From the leaves of *Ocotea glaziovii* have been isolated 3,5-dihydroxy-6-methoxyaporphine (I) and the isoquinolinedienone alkaloid glaziovine (III). The dienone–phenol rearrangement converts glaziovine to I, while by successive borohydride reduction and rearrangement, it is transformed to 5-demethylnuciferine (V).

The large and widely spread genus *Ocotea* of the family *Lauraceae* has already been the subject of considerable chemical study both as a source of alkaloids of the benzylisoquinoline group and of essential oils.[6] Although some 120 Brazilian species have been reported, chemical investigations seem to have been limited to a study of nonalkaloidal components.[6b] We therefore thought it of interest to study the alkaloids of *Ocotea glaziovii* Mez, collected in the Tijuca Forest of Rio de Janeiro.

Examination of the leaves provided two principal isomeric alkaloids of empirical formula $C_{18}H_{19}NO_3$. One of these, glaziovine, was isolable by chloroform

extraction from an acetic or tartaric acid solution of the total bases at pH 4–5, while the other base was only extracted by organic solvents after alkalinization of the aqueous solution. This latter base showed ultraviolet absorption typical of an aporphine oxygenated at positions 3, 5, and 6[7a,8] (see Experimental section) and was identified as (−)-3,5-dihydroxy-6-methoxyaporphine (I).[7a,b] Aporphine I was isolated as its hydrochloride and characterized as its methochloride monohydrate, prepared from the methiodide. Both the free base I and its methiodide suffer ready air oxidation. The n.m.r. spectrum was similar to that of tuduranine (3-hydroxy-5,6-dimethoxy-N-demethylaporphine, II)[9] as shown in Table I, the additional down-

(7) (a) See T. Kitamura, *J. Pharm. Soc., Japan*, **80**, 1104 (1960), who obtained 3,5-dihydroxy-6-methoxyaporphine (I) in very low yield from the oil resulting from the reduction of domesticine with sodium in liquid ammonia, and characterized it as its hydrochloride, m.p. 277° dec. (b) The negative rotation of I points to the *R*-absolute configuration [C. Djerassi, K. Mislow, and M. Shamma, *Experientia*, **18**, 53 (1962); M. Shamma, *ibid.*, **18**, 64 (1962)] and this was confirmed by measurement of the rotatory dispersion of V.

(8) L. J. Haynes and K. L. Stuart [*J. Chem. Soc.*, 1789 (1963)] describe a number of aporphine derivatives oxygenated in these three positions (see ref. 16).

field shift of the C_4-proton indicating an oxygen function in position 5.[10,11] The aporphine I did not show catecholic properties[12] and the methoxyl group therefore could be placed at C-6. This placement also is consistent with the O-methyl assignments of Table I. Methylation of I with dimethyl sulfate gave 3,5,6-trimethoxyaporphine methosulfate, identical by infrared comparison with O,N-dimethyltuduranine methosulfate prepared from tuduranine (II).[7]

TABLE I

N.m.r. Spectra of Aporphines I and V and Tuduranine

			δ-Values		
		I	Tuduranine, II		V
Group	No. of protons	a	a	b	b
N–CH₃	3, singlet	2.57	..	..	2.5
C₅–OCH₃	3, singlet	..	3.62	3.62	...
C₆–OCH₃	3, singlet	3.92	3.75	3.82	3.8
OH–NH	1–2, singlet	..	..	5.16	5.7
C₁–H	1, doublet^c	7.18	6.98	6.94	$(7.1–7.4^d)$
C₁–H	1, quartet^c	6.62	6.63	6.57	
C₄–H	1, doublet^c	8.08	7.65	7.86	$(8.2–8.4^e)$
C₇–H	1, singlet	6.77	6.57	6.52	6.55

^{a} Measured in D_2O–NaOD at 60 Mc. using benzene as an external standard; δ-values corrected to SiMe₄; $\delta = 0$, $C_6H_6 = 295$ c.p.s. ^{b} Measured in $CDCl_3$ at 60 Mc. using SiMe₄ as an internal standard. ^{c} J_{HH} for C_1–H = 7.8; for C_2–H, C_4–H = 2.5 c.p.s. in I; for C_1–H, C_2–H = 8.8; for C_2H, C_4–H = 2.3 c.p.s. in II^a; for C_1–H, C_2–H = 8.3; for C_2–H, C_4–H = 1.9 c.p.s. in II.^{b} ^{d} Multiplet corresponding to the 3 protons on C_1, C_2, C_3. ^{e} Multiplet.

Glaziovine, which is relatively stable to aerial oxidation, was characterized as its picrate. The free base shows infrared absorption attributable to an α,β-unsaturated ketone, and it forms an oxime. On hydrogenation over platinum in ethanol at room temperature, 2 moles of hydrogen was taken up to give tetrahydroglaziovine (IV), whose infrared absorption was indicative of a saturated six- or larger membered ring ketone. The ultraviolet spectrum of the tetrahydro derivative (see Table II) was comparable to that of

TABLE II

Ultraviolet Absorption Spectra of Glaziovine and Tetrahydroglaziovine

|	In ethanol		In ethanolic KOH	
	λ_{max}, mμ	ϵ	λ_{max}, mμ	ϵ
Glaziovine	288	3710	308	5440
Tetrahydroglaziovine	286	2610	253	6120
			299	4450
2-Methoxy-3,4,5-trimethyl-phenol	284	2340	295	3980

2-methoxy-3,4,5-trimethylphenol. Glaziovine itself showed similar but more intense absorption, not incompatible with the superimposition of a cyclohexadienone chromophore upon that of the above-mentioned trimethylguaiacol, if some interaction were to occur between the p-electrons of the phenolic oxygen with the π-electrons of the dienone. In accordance with the foregoing evidence, glaziovine undergoes the dienone–phenol rearrangement with cold hydrochloric acid to give the $(-)$-aporphine I. The structures III and IV therefore could be proposed for the alkaloid and its tetrahydro derivative, and these proved to

(9) We are indebted to Professor K. Goto for a generous gift of tuduranine; cf. *Ann.*, **521**, 175 (1936).

(10) S. Goodwin, J. N. Shoolery, and L. F. Johnson, *Proc. Chem. Soc.*, 306 (1958).

(11) I. R. C. Bick, J. Harley-Mason, N. Sheppard, and M. J. Vernengo, *J. Chem. Soc.*, 1896 (1961).

(12) S. McLean, K. Palmer, and L. Marion, *Can. J. Chem.*, **38**, 1547 (1960).
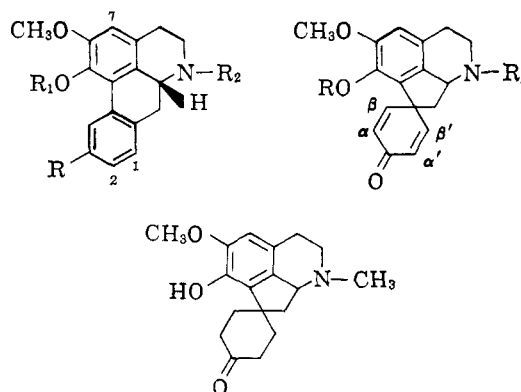
I, R = OH, R₁ = H, R₂ = CH₃   III, R = H, R₁ = CH₃
II, R = OH, R₁ = CH₃, R₂ = H   VII, R = R₁ = CH₃
V, R = R₁ = H, R₂ = CH₃   VIII, R = R₁ = H
VI, R = H, R₁ = R₂ = CH₃

IV

be in full accord with the n.m.r. data reported in Table III.

Sodium borohydride reduction of glaziovine (III) gave an alcohol which readily underwent elimination of the elements of water and rearrangement on treatment with cold dilute hydrochloric acid during isolation to give R-5-hydroxy-6-methoxyaporphine (V),[7b] which was identified by its ultraviolet and n.m.r. spectra (Table I). Methylation of V with diazomethane gave 5,6-dimethoxyaporphine, identical with an authentic specimen of nuciferine (VI).[13]

The hydroxyl group eliminated is thus that which gives rise to the phenolic function at C-3 of the aporphine I, confirming structure III for glaziovine. Glaziovine thus represents a third member of a new group of isoquinolinedienone alkaloids[14] of which pronuciferine (VII)[15,16] and crotonosine (VIII)[16] are members, differing from glaziovine only in degree of methylation. Fungapavine[17] may be another example of this class.

Experimental[18]

Isolation of Alkaloids from *Ocotea glaziovii* **Mez.**—The dried ground leaves (9.2 kg.) were digested with 95% ethanol and the ethanol was removed by distillation at atmospheric and finally reduced pressure to give a resinous extract (1615 g.). Portions

(13) We wish to thank Professors H. R. Arthur and R. G. Cooke for authentic specimens of nuciferine; cf. H. R. Arthur and H. T. Cheung, *J. Chem. Soc.*, 2306 (1959); R. G. Cooke and H. F. Haynes, *Australian J. Chem.*, **7**, 99 (1954). Natural nuciferine is levorotatory, having the *R*-configuration (see footnote 7b).

(14) This type of structure has been postulated by D. H. R. Barton and T. Cohen, "Festschrift Arthur Stoll," 1957, p. 124, as a possible biogenetic precursor of aporphines and other alkaloids.

(15) K. Bernauer, *Helv. Chim. Acta*, **46**, 1783 (1963).

(16) Reference 15, footnote 5a; L. J. Haynes and K. L. Stuart, *J. Chem. Soc.*, 1784, 1789 (1963); L. J. Haynes, K. L. Stuart, D. H. R. Barton, and G. W. Kirby, *Proc. Chem. Soc.*, 280 (1963).

(17) Fungapavine, an α,β-unsaturated ketonic alkaloid isolated by V. A. Mnatsakanyan and S. Yu. Yunusov [*Dokl. Akad. Nauk. Uz. SSR*, **No. 12**, 36 (1961)], was found to isomerize on treatment with acid to an aporphine. It was assigned the highly unlikely structure i, which would certainly exist entirely in the enolic aporphine form. An interesting alternative for fungapavine is ii, consistent with its reported properties.
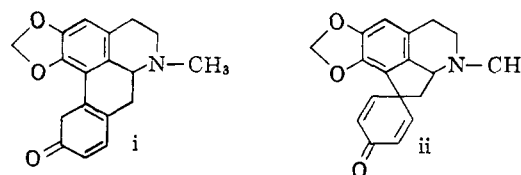
(18) Melting points are uncorrected. Analyses were performed by Dr. A. Bernhardt, Mülheim, Germany, and the Microchemical Laboratory, University of California, Berkeley. Purity of all substances was determined by chromatography on Whatman No. 1 paper or silica gel chromatoplates using in both cases butanol–acetic acid–water mixtures as developing solvents.

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TABLE III

N.M.R. Spectra of Glaziovine and Tetrahydroglaziovine

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| | | ‑‑‑‑‑‑δ-Values[a]‑‑‑‑‑‑ | |
Group	No. of protons	Glaziovine	Tetrahydro-glaziovine
N–CH₃	3, singlet	2.4	2.35
O–CH₃	3, singlet	3.85	3.75
OH	1, broad singlet	6.0	4.5–5.3
Aromatic proton	1, singlet	6.65	6.5
Vinyl protons	4, two AB-type	6.25–6.6(α,α')	...
	quartets with	6.7–7.3(β,β')	...
	fine structure[b]		

[a] Measured at 60 Mc. in CDCl₃; values relative to SiMe₄ as an internal standard, $\delta = 0$. [b] The α- and α'-protons show marked interaction across the ring; this is not observed in the case of β,β'-protons.

of this extract (100 g.) were dispersed in dilute aqueous tartaric acid (pH 4, 1 l.) and filtered, and the filtrate was extracted continuously with chloroform. The chloroform solution on evaporation yielded 1–2 g. of crude alkaloid which, on recrystallization from ether, ether–methanol, and ethyl acetate, gave **glaziovine (III)** as colorless needles, m.p. 235–237° dec., $[\alpha]_D +7°$ (c 1.0, chloroform); $\nu_{max}^{CHCl_3}$ 1657 (s), 1619 (s) cm.$^{-1}$.

Anal. Calcd. for C₁₈H₁₉NO₃: C, 72.7; H, 6.4; N, 4.7; 1 OCH₃, 10.4; mol. wt., 297. Found: C, 72.4; H, 6.2; N, 5.0; OCH₃, 10.5; mol. wt., 297, mass spec.[19]

The picrate crystallized from ethanol and had m.p. 199–203°.

Anal. Calcd. for C₁₈H₁₉NO₃·C₆H₃N₃O₇: N, 10.4. Found: N, 10.6.

Further chloroform extractions of the above aqueous solution at pH 5 and 6 resulted in the isolation of further small amounts of glaziovine. The aqueous solution was then made alkaline with ammonium hydroxide and extracted with ether. Evaporation of the ethereal extract gave crude **3,5-dihydroxy-6-methoxyaporphine (I)**, but as this suffered ready air oxidation, a preferred method of isolation involved the addition of a few drops of concentrated hydrochloric acid to the ethereal extract, which resulted in the precipitation of the hydrochloride, dec. >300°, from which the free base I was isolated as colorless crystals of monohydrate (0.5 g.), m.p. 149–152° dec., $[\alpha]^{26}_D -35°$ (c 0.2, chloroform); ν_{max}^{Nujol} 3460 (w), 3300 (m), 1600 (m), 1302 (m), 1136 (m) cm.$^{-1}$; λ_{max}^{EtOH} 218, 266, 275, and 307 mμ, ε 38100, 10200, 13400, 9040; $\lambda_{max}^{EtOH\ -NaOH}$ 340 mμ, ε 9590[7]; neutral spectrum unchanged by the presence of boric acid buffered with sodium acetate.[12]

Anal. Calcd. for C₁₈H₁₉NO₃·H₂O: C, 68.6; H, 6.7; N, 4.4; 1 OCH₃, 9.8. Found: C, 68.9; H, 6.5; N, 4.2; OCH₃, 9.8.

(19) The mass spectrum of glaziovine was measured by C. Djerassi and H. Budzikiewicz, Stanford University, and showed peaks at the following positions, M⁺, 297; M − 17; M − 29; M − 43; M − 58; M − 86; m/e 165.

Compound I did not show fluorescence in ultraviolet light when treated with 10% aqueous ethylenediamine,[20] demonstrating the absence of a catechol grouping.

With methyl iodide, compound I gave a crystalline methiodide, m.p. 251–253°, which darkened rapidly in air. A solution of the methiodide in aqueous methanol was stirred with freshly prepared silver chloride for 1 hr. and filtered.[21] The filtrate was evaporated and the residue recrystallized from methanol–acetone giving I·methochloride·hydrate, m.p. 226–229°.

Anal. Calcd. for C₁₉H₂₂ClNO₃·H₂O: C, 62.4; H, 6.6. Found: C, 62.4; H, 6.8.

3,5,6-Trimethoxyaporphine Methosulfate.—3,5-Dihydroxy-6-methoxyaporphine (I, 226 mg.) was boiled during 72 hr. with dimethyl sulfate (0.1 ml.) in acetone (10 ml.) in the presence of anhydrous potassium carbonate. The mixture was filtered and the filtrate concentrated, whereupon 3,5,6-trimethoxyaporphine methosulfate (172 mg.) crystallized. The salt was recrystallized from acetone–hexane; m.p. 189–202° dec.; ν_{max}^{KCl} 1608, 1582, 1506, 1420 cm.$^{-1}$, identical with a sample prepared similarly from tuduranine (II).[9]

Tetrahydroglaziovine (IV).—Glaziovine (100 mg.) was hydrogenated in acetic acid (10 ml.) in the presence of platinum oxide (50 mg.), until two molar proportions of hydrogen had been absorbed. The solution was filtered and evaporated and the crude **tetrahydroglaziovine** (96 mg.), m.p. 105–120°, crystallized from benzene to give colorless crystals, m.p. 112–116°; $\nu_{max}^{CHCl_3}$ 3520, 1699 cm.$^{-1}$.

Anal. Calcd. for C₁₈H₂₁NO₃·¹/₆C₆H₆: C, 73.1; H, 7.1; N, 4.5. Found: C, 73.3; H, 7.6; N, 4.3.

5-Hydroxy-6-methoxyaporphine (V).—Glaziovine (100 mg.) was stirred in 50% aqueous methanol (10 ml.) with sodium borohydride (130 mg.) during 45 min. The solution was then acidified with diluted hydrochloric acid, alkalinized with ammonia, and extracted with ether. Addition of a drop of concentrated hydrochloric acid resulted in the slow crystallization of 5-hydroxy-6-methoxyaporphine hydrochloride hemihydrate (116 mg.), dec. >220°; ν_{max}^{Nujol} 1610 (m), 1577 (w), 1499 (m), 781 (m), 752 (s) cm.$^{-1}$.

Anal. Calcd. for C₁₈H₂₀ClNO₂·¹/₂H₂O: C, 66.2; H, 6.2. Found: C, 66.7; H, 6.0.

The free base V liberated with ammonia had m.p. 167–169°; R.D.[22] in dioxane (c 0.17): $[\alpha]_{589} -150°$, $[\alpha]_{400} -360°$, $[\alpha]_{340} -560°$, $[\alpha]_{318} -600°$, $[\alpha]_{310} -520°$, $[\alpha]_{305} 0°$, $[\alpha]_{300} +2560°$, $[\alpha]_{285} +7680°$, $[\alpha]_{270} 0°$, $[\alpha]_{250} -22,940°$; λ_{max}^{EtOH} 271 and 310 mμ, ε 13700, 3900; $\lambda_{max}^{EtOH-KOH}$ 270 and 345 mμ, ε 7100, 4850. Methylation of V (10 mg.) with diazomethane in ether–methanol during 2 weeks followed by evaporation of the solution and purification of the product by thin layer chromatography gave nuciferine (5,6-dimethoxyaporphine, VI) identical with an authentic sample[13] by infrared and chromatographic comparison.

(20) H. Weil-Malherbe and A. D. Bone, *Biochem. J.*, **51**, 311 (1952).

(21) M. Tomita and I. Kikkawa, *Pharm. Bull. Japan*, **4**, 230 (1956).

(22) Measured by C. Djerassi and R. Records with an automatically recording spectropolarimeter.

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An Experimental Assignment of the Proton Magnetic Resonance Spectrum of Purine[1]

By M. P. Schweizer, Sunney I. Chan,[2] G. K. Helmkamp, and P. O. P. Ts'o

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The assignment of the proton n.m.r. spectrum of purine has been made on the basis of specific deuteration procedures. In aqueous solution, the spectrum consists of three lines. The high field peak was shown to arise from the 8-proton on the imidazole ring, for desulfurization of 8-mercaptopurine with deuterated Raney nickel, or exchange of that proton with deuterium resulted in the decrease of intensity of that peak. The low field peak has been assigned to the 6-position of the pyrimidine ring by a similar desulfurization of 6-mercaptopurine and by reduction of 6-iodopurine with Adams catalyst and deuterium in methanol-d. The middle peak was assigned to the 2-proton on the pyrimidine ring by difference. This assignment of the proton n.m.r. spectrum of purine can be rationalized on the basis of an improved theory relating π-electron densities to proton chemical shifts in aromatic systems.

Introduction

In an investigation of the association of purine in aqueous solution,[3] we have been concerned with the

(1) This work was supported in part by the Research Corporation, and in part by Grants GM-08185-03, RG-5143, GM-10316-01 from the National Institutes of Health, U. S. Public Health Service.

(2) Gates and Crellin Laboratories of Chemistry, California Institute of Technology, Pasadena, Calif.

assignment of the protons observed in its n.m.r. spectrum. The actual assignment was critical for the interpretation of the solute–solute interaction. Several modes of interaction were conceivable, and n.m.r. spectral results could be properly assessed in terms of a

(3) S. I. Chan, G. K. Helmkamp, M. P. Schweizer, and P. O. P. Ts'o, paper presented at the Symposium on Molecular Structure and Spectroscopy, Columbus, Ohio, June 10–14, 1963.