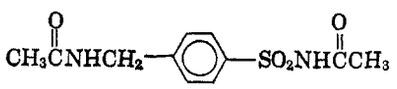
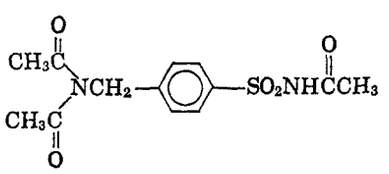
We have re-examined the reaction and have found that the two "tautomers" are actually the $N^1,N^4$-diacetyl (mp 214°) II and $N^1,N^4,N^4$-triacetyl (mp 196°) III derivatives. Reaction of $N^4$-acetyl-4-(aminomethyl)benzenesulfonamide with 1 mole of acetic anhydride gave the $N^1,N^4$-diacetyl compound in 99% yield. Increasing the anhydride/sulfonamide molar ratio to three resulted in the isolation of the triacetyl and diacetyl compounds in an 11:3 ratio (over-all 70% yield). When 4-(aminomethyl)benzenesulfonamide was treated with 8 moles of anhydride, only the triacetyl derivative was obtained (68% yield).



II



III

The position of the third acetyl group was ascertained by inspection of the infrared and pmr spectra. Transformation of the methylene doublet to a singlet with a concomitant downfield shift of $\delta$ 0.67 unambiguously places the third acetyl group on the benzylamine nitrogen. This assignment is confirmed by the absence of the 6.1-$\mu$ carbonyl band (shown by the $N^4$-acetyl and $N^1,N^4$-diacetyl derivatives) and the presence of two new carbonyl bands at 5.80 and 5.94 $\mu$ in the infrared spectrum of the triacetyl compound[2] (Table I).

TABLE I

SPECTRA OF ACETYLATED 4-(AMINOMETHYL)BENZENESULFONAMIDES

| Derivative of 4-(aminomethyl)-benzenesulfonamide | Mp, °C | Pmr, $\delta$[a] | Infrared C=O $\mu$[b] |
|---|---|---|---|
| $N^4$-Acetyl | 175–176.5[c] | 1.93 (3, s), 4.37 (2, d) | 6.1 |
| $N^1,N^4$-Diacetyl | 214–216 | 1.93 (3, s), 1.95 (3, s), 4.37 (2, d) | 5.84, 6.1 |
| $N^1,N^4,N^4$-Triacetyl | 195–196 | 2.37 (6, s), 1.95 (3, s), 5.03 (2, s) | 5.8, 5.84, 5.94 |

[a] With respect to internal standard of tetramethylsilane in deuterated dimethyl sulfoxide; spectra obtained on a Varian A-60 spectrometer. Number of protons is in parentheses: s, singlet; d, doublet. [b] KBr disks. [c] J. Klarer [U. S. Patent 2,288,531 (June 30, 1942); *Chem. Abstr.* **37**, 888 (1943)] reported mp 177°.

(1) M. Ishidate and T. Momose, *J. Pharm. Soc. Japan*, **67**, 214 (1947); *Chem. Abstr.*, **45**, 8994*a* (1951).

(2) Cyclic diacylimides show two carbonyl bands for apparently equivalent moieties (H. M. Randall, R. G. Fowler, N. Fuson, and J. Dangl, "Infrared Determination of Organic Structures," Van Nostrand Co., Inc., New York, N. Y., 1949, pp 14, 20). R. A. Abramovitch [*J. Chem. Soc.*, 1413 (1957)] has demonstrated that symmetrically substituted N,N-diacetylanilines reported [J. F. Grove, P. W. Jeffs, and D. W. Rustidge, *J. Chem. Soc.*, 1956 (1956)] to possess only one carbonyl band do in fact on refinement of experimental technique (slit schedules narrowed to 0.5 mm) possess two bands in the carbonyl region. T. Uno and K. Machida [*Bull. Chem. Soc. Japan*, **34**, 545 (1961)] have reported that diacetamide exists in two forms, the "A" *cis–trans* form (relative to the nitrogen proton) in which the compound is usually found showing two carbonyl bands in mulls. Unpublished X-ray data (T. Watanabé and K. Osaki) is cited as establishing the geometric isomerism. C. M. Lee and W. D. Kumler [*J. Am. Chem. Soc.*, **84**, 571 (1962)] have from dipole moment data reached the same conclusion. The less stable "B" form, considered by Uno and Machida to be the *trans–trans* isomer, displays only one major carbonyl band (slight shoulders excluded, slit schedule not specified).

(3) K. B. Sandell, *Monatsh. Chem.*, **92**, 1066 (1961).

(4) N. Bacon, A. J. Boulton, R. T. Brownlee, A. R. Katritzky, and R. D. Topsom, *J. Chem. Soc.*, 5230 (1965).

(5) We thank W. Fulmor and associates for the spectra and L. Brancone and associates for the microanalytical data. Melting points were determined in a Mel-Temp apparatus and are corrected. $R_f$ values are for descending partition paper chromatography in *n*-BuOH–concentrated $NH_4OH$–$H_2O$ 9:1:8.

(6) See footnote *c* of Table I.

The triacetylsulfonamide was found to be readily hydrolyzed in dilute base at room temperature to the $N^1,N^4$-diacetyl compound. Therefore, the interconversion of A and B is merely a hydrolysis–reacetylation process.

It is of interest to note that, not surprisingly, the claim of Sandell[3] that primary benzenesulfonamides exist partially or predominantly in the imido form has been demonstrated recently by Katritzky and coworkers[4] to be incorrect.

### Experimental Section[5]

**Preparation of $N^1,N^4$-Diacetyl-4-(aminomethyl)benzenesulfonamide.**—A mixture of 6.0 g of 4-(N-acetylaminomethyl)benzenesulfonamide[6] ($R_f$ 0.66) and 2.65 g of acetic anhydride in 15 ml of pyridine was refluxed for 2 hr. The pyridine was removed by evaporation in a stream of air and the residue was dried in a vacuum oven at 70–80°. The white solid, 7.05 g (99%), melted at 207–211°. Crystallization from ethanol gave a sample melting at 211–213° ($R_f$ 0.21).

*Anal.* Calcd for $C_{11}H_{14}N_2O_4S$: C, 48.9; H, 5.2; N, 10.4; S, 11.9. Found: C, 48.9; H, 5.2; N, 10.2; S, 11.7.

**Mixture of $N^1,N^4$-Diacetyl and $N^1,N^4,N^4$-Triacetyl Derivatives.**—Acetic anhydride (6.5 g) and 4.6 g of 4-(N-acetylaminomethyl)-benzenesulfonamide were refluxed for 2 hr. The addition of 40 ml of water to the cooled solution produced an oil which soon crystallized to give 3.5 g of white solid, mp 185–189° (55%). Recrystallization of 0.6 g from 40 ml of ethanol yielded 0.37 g, mp 195–196°, of the triacetyl derivative.

*Anal.* Calcd for $C_{13}H_{16}N_2O_5S$: C, 50.0; H, 5.2; N, 9.0; S, 10.3. Found: C, 50.5; H, 5.2; N, 8.8; S, 10.3.

On standing, the filtrate of the 3.5 g of crop yielded 0.8 g (15%) of material melting over a range of 170–185° and possessing an infrared spectrum superimposable upon that of $N^1,N^4$-diacetyl-4-(aminomethyl)benzenesulfonamide.

**Preparation of $N^1,N^4,N^4$-Triacetyl Derivative from 4-(Aminomethyl)benzenesulfonamide and Hydrolysis to $N^1,N^4$-Diacetyl Derivative.**—A solution of 4.65 g of 4-(aminomethyl)benzenesulfonamide, 2.05 g of sodium acetate, and 20.4 g of acetic anhydride was refluxed for 2 hr. The addition of 125 ml of water produced 5.2 g of the triacetyl compound, mp 196–198°, yield 65%.

This material (0.5 g) was dissolved in 10 ml of 0.4 *N* NaOH solution and allowed to stand at room temperature for 1.5 hr. Adjustment of the pH to 3.5 with 1 *N* HCl solution resulted in the precipitation of 0.33 g of $N^1,N^4$-diacetyl-4-(aminomethyl)-benzenesulfonamide, mp 214–216°.

## Tetrahydro-2-pyranyl Derivatives of Purines

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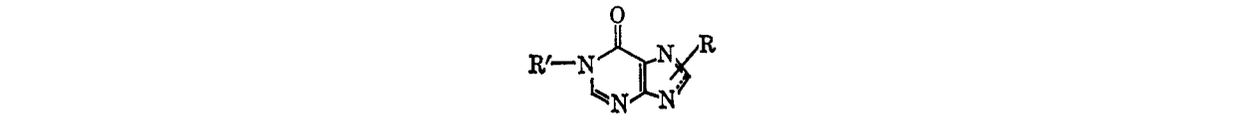
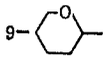
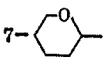
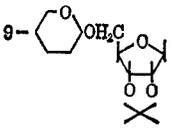
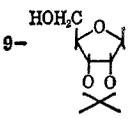
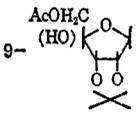
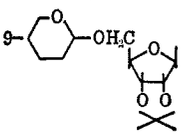
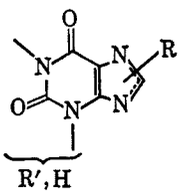
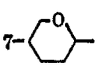
*Received January 11, 1966*

Robins and co-workers[1] have reported that certain 6-substituted purines react with 2,3-dihydro-4H-pyran

(1) R. K. Robins, E. F. Godefroi, E. C. Taylor, L. R. Lewis, and A. Jackson, *J. Am. Chem. Soc.*, **83**, 2574 (1961). L. R. Lewis, F. H. Schneider, and R. K. Robins, *J. Org. Chem.*, **26**, 3837 (1961). W. A. Bowles, F. H. Schneider, L. R. Lewis, and R. K. Robins, *J. Med. Chem.*, **6**, 471 (1963).

TABLE I

ULTRAVIOLET ABSORPTION SPECTRA OF CERTAIN SUBSTITUTED PURINES

| R | R′ | | pH 1 $\lambda_{max}$ | pH 1 $\epsilon \times 10^{-3}$ | pH 11 $\lambda_{max}$ | pH 11 $\epsilon \times 10^{-3}$ | Ethanol $\lambda_{max}$ | Ethanol $\epsilon \times 10^{-3}$ |
|---|---|---|---|---|---|---|---|---|
|  | | | | | | | | |
| 9-  |  | II | 250 | 10.9 | 251.5<br>(270) | 10.1 | 246<br>251<br>(270) | 9.3<br>9.1 |
| 7-  |  | III | 250 | 10.0 | 257<br>(270) | 7.0 | 258<br>(270) | 6.6 |
| 9-  | H | VII | 252 | 8.6 | 254.5 | 12.7 | 246<br>250<br>(267) | 11.2<br>11.1 |
| 9-Ribofuranosyl | H | | 252 | | 254 | 13.5 | 246<br>250<br>(267) | 11.1<br>11.1 |
| 9-Benzyl | Me | | 252[a] | 11.3 | 252[a] | 11.3 | | |
| 7-Benzyl | Me | | 255[a] | 9.8 | 258[a] | 9.1 | | |
| 9-  | H | | 251.5 | | 253.5 | 13.3 | 246<br>250<br>(267) | 11.2<br>11.1 |
| 9-  | Me | IV | 254.5 | 9.5 | 251<br>(270) | 9.1 | 246<br>251<br>(270) | 8.0<br>7.7 |
| 9-  | Me | VIII | 254.5 | 9.1 | 251.5<br>(270) | 8.6 | 246<br>251<br>(270) | 7.9<br>7.9 |
|  | | | | | | | | |
| 7-  | H | VI | (232)<br>262 | 8.6 | (234)<br>292.5 | 7.8 | 272.5 | 8.1 |
| 9-Ribofuranosyl | H | | (232)<br>262 | | 243<br>278.5 | | 268 | |
| 9-Me | H | | | | 277[b] (pH 10) | | | |
| 7-Me | H | | | | 287[b] | | | |
| H | 1-Me | | | | 242[b] (pH 10)<br>276[b] (pH 10) | | | |
| H | 3-Me | | | | 274[b] (pH 10) | | | |

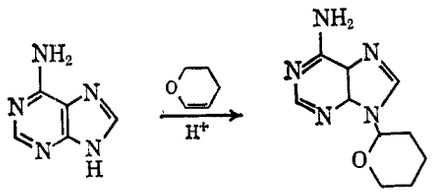
[a] L. B. Townsend and R. K. Robins, *J. Org. Chem.*, **27**, 990 (1962). [b] Reference 5.

in ethyl acetate in the presence of *p*-toluenesulfonic acid to give 9-(tetrahydro-2-pyranyl) derivatives. This reaction was applied successfully by them to 6-chloro-, -iodo-, -methoxy-, or -methylthiopurines, but not to adenine, hypoxanthine, purine, or 6-purinethiol. This paper is concerned with the problem involved in the introduction of a tetrahydro-2-pyranyl group directly into naturally occurring purines.

Adenine was treated with 2,3-dihydro-4H-pyran in dimethyl sulfoxide at 55–60° in the presence of a small excess of hydrogen chloride to produce tetrahydro-2-pyranyladenine (I) in good yield. The infrared absorption spectrum of I shows two strong characteristic bands of tetrahydropyran structure at 1082 and 1043 $cm^{-1}$.[2] The tetrahydro-2-pyranyl group of I was assigned to position 9 by virtue of the similarity of the ultraviolet absorption spectrum of I ($\lambda_{max}$ 261 m$\mu$ at pH 11) to that for adenosine ($\lambda_{max}$ 261 m$\mu$ at pH 11) and 9-methyladenine ($\lambda_{max}$ 260 m$\mu$ at pH 11).[3] On the other hand, the spectrum of I differs from those for 7- and 3-methyladenines ($\lambda_{max}$ 270 and 273 m$\mu$, respectively, at pH 11).[3] Robins, *et al.*,[1] have obtained

(2) A. J. Speziale, K. W. Ratts, and G. J. Marco, *J. Org. Chem.*, **26**, 4311 (1961). They reported three bonds characteristic of tetrahydropyran, but the middle of those is very weak.

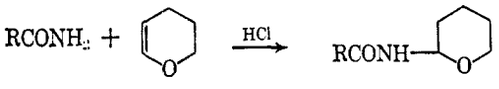
(3) N. J. Leonard and J. A. Deyrup, *J. Am. Chem. Soc.*, **84**, 2148 (1962).



I

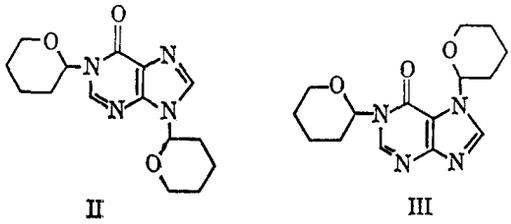
I by treatment of 6-chloro-9-(tetrahydro-2-pyranyl)-purine with ammonia and have found essentially similar properties for I ($\lambda_{max}$ 262 mμ, ε 13,900 in ethanol).

Similarly, di-(tetrahydro-2-pyranyl)hypoxanthine (II) (mp 183–184°) and a small amount of its isomer (III) (mp 160–162°) were obtained from hypoxanthine. The tetrahydro-2-pyranyl groups of II was assigned to position 1 and 9 by comparison of the ultraviolet absorption spectra of II with those of 1-methyl-5′-O-acetyl-2′,3′-O-isopropylideneinosine (IV) prepared from 5′-O-acetyl-2′,3′-O-isopropylideneinosine (V) with diazomethane (Table I). IV was hydrolyzed with acid to produce 1-methylhypoxanthine, whose $R_f$ values and the ultraviolet absorption spectrum both were identical with those reported by Miles.[4] The infrared absorption spectrum of II shows a strong absorption band at 1694 $cm^{-1}$ presumably due to C=O stretching vibration. An analogous reaction has been reported by Speziale, Ratts, and Marco;[2] various amides react with 2,3-dihydro-4H-pyran to give N-(tetrahydro-2-pyranyl) derivatives. These facts support the assign-

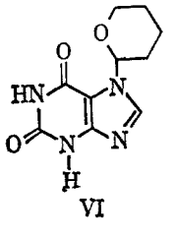


R = $C_6H_5$–, $CH_3$–, $(CH_3)_2CH$–, etc.

ment of one of the tetrahydro-2-pyranyl groups to position 1. Compound III, the isomer of II, has been assumed to be 1,7-di-(tetrahydro-2-pyranyl)hypoxanthine, since the ultraviolet absorption spectrum of III is similar to 1-methyl-7-benzylhypoxanthine (Table I), and an infrared absorption spectrum shows a strong absorption band at 1692 $cm^{-1}$.



II III

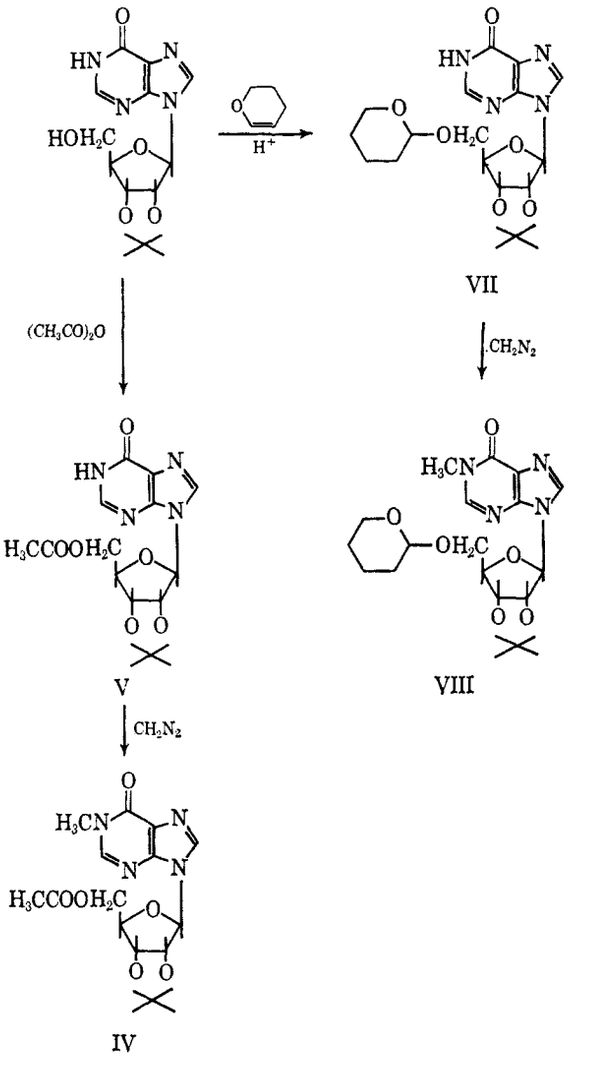


VI

Tetrahydro-2-pyranylxanthine (VI) was the main product from a similar reaction of xanthine with 2,3-dihydro-4H-pyran. Di- and tri(tetrahydro-2-pyranyl)-xanthine were also obtained in very low yields and were not investigated further. The ultraviolet absorption spectrum of VI at pH 11 is markedly different from that of xanthosine (Table I) and exhibits an absorption maximum at 292.5 mμ (*cf.* xanthosine, 278.5 mμ). Pfleiderer, *et al.*,[5] have shown that the ultraviolet absorption spectrum of 7-methylxanthine at pH 11 exhibits an absorption maximum at 287 mμ, while those of 1-methyl-, 3-methyl- and 9-methylxanthine appear at 276, 274, and 277 mμ, respectively. VI is assumed, therefore, to be 7-(tetrahydro-2-pyranyl)-xanthine.

Some attempts were made unsuccessfully to prepare tetrahydro-2-pyranyl derivatives of uracil and cytosine.

Reaction of 2,3-dihydro-4H-pyran with 5′-O-acetyl-2′,3′-O-isopropylideneinosine (V) did not give rise to the formation of any tetrahydro-2-pyranyl derivative but rather to an oily product together with a half of the unchanged starting material. However, 2′,3′-O-isopropylideneinosine reacted with 2,3-dihydro-4H-pyran to give a tetrahydro-2-pyranyl derivative (VII).



The absorption bands at 1702 and near 3100 $cm^{-1}$ in the infrared absorption spectrum of VII show the presence of C=O and NH groups. The ultraviolet absorption spectrum of VII at pH 11 shows a maximum at 254.5 mμ similar to inosine, 2′,3′-O-isopropylidene-

(4) H. Bredereck and A. Martini, *Chem. Ber.*, **80**, 401 (1947). H. T. Miles, *J. Org. Chem.*, **26**, 4761 (1961).

(5) W. Pfleiderer and G. Nubel, *Ann.*, **647**, 155 (1961).

inosine, and 5′-O-acetyl-2′,3′-O-isopropylideneinosine (V) at 254, 253.5, and 253.5 mμ, respectively. These results lead to the assignment of the tetrahydro-2-pyranyl group of VII to the 5′ position of sugar moiety. Further structural proof was obtained by conversion of VII and V to their corresponding methyl derivatives (VIII, IV), respectively, by treatment with diazomethane. The ultraviolet absorption maxima of both methyl derivatives shifted to shorter wave length, 251–251.5 mμ. No band was found near the 3100-$cm^{-1}$ region in the infrared spectrum of VIII which might be assigned to NH vibration. On the basis of the infrared and ultraviolet absorption spectra, it is concluded that the methyl group was situated on N-1 in purine ring. These facts support the structure of VII as 5′-O-(tetrahydro-2-pyranyl)-2′,3′-O-isopropylideneinosine.

### Experimental Section

Paper chromatography was carried out in the following solvent systems: solvent A, *n*-butyl alcohol–acetic acid–water (32:15:8); solvent B, isopropyl alcohol–concentrated ammonium hydroxide–water (7:1:2).

**9-(Tetrahydro-2-pyranyl)adenine (I).**—Ten grams of adenine (0.074 mole) was dissolved in 100 ml of dimethyl sulfoxide with 12 ml of a solution of hydrogen chloride in dry dioxane (7 *N*). Twenty ml of 2,3-dihydro-4H-pyran was added to the stirred solution at 55°–60°. The mixture was kept at 55–60° for 2 hr. A white solid (15 g) precipitated and, after filtration, was dissolved in a small amount of water. The aqueous solution was adjusted to pH 7 and cooled to give 10 g (62%) of crude I. Recrystallization from ethyl acetate gave colorless crystals, mp 181–181.5°.

*Anal.* Calcd for $C_{10}H_{13}N_5O$: C, 54.78; H, 5.98; N, 39.95. Found: C, 54.41; H, 6.26; N, 39.86.

**1,9-Di(tetrahydro-2-pyranyl)hypoxanthine (II).**—Eight grams of hypoxanthine (0.058 mole) was dissolved in 100 ml of dimethyl sulfoxide with 10 ml of a solution of hydrogen chloride in dry dioxane (7 *N*) with stirring at 55–60°. 2,3-Dihydro-4H-pyran (40 ml) was added to the solution and the mixture was held at 55–60° for 15 hr. No precipitate was appeared. After 7 ml of concentrated aqueous ammonium hydroxide was added, the mixture was concentrated under reduced pressure. The resulting gum was dissolved in ethyl acetate and the solution was washed with water. The organic layer was dried over anhydrous sodium sulfate. The solvent was evaporated under reduced pressure, and the residue was dissolved in ethyl acetate. A white precipitate (8 g, 45%) formed. Recrystallization from ethyl acetate gave II, colorless crystals: mp 183–184°; $R_f$ 0.92 (A), 0.96 (B). The mother liquor was saved for the next step.

*Anal.* Calcd for $C_{15}H_{20}N_4O_3$: C, 59.19; H, 6.62; N, 18.41. Found: C, 59.29; H, 6.67; N, 18.66.

**1,7-Di(tetrahydro-2-pyranyl)hypoxanthine (III).**—Concentration of the mother liquor of II left a residual syrup which was dissolved in benzene, applied to an alumina column and eluted with benzene–ethanol (ethanol: 0, 1, 5, 10, 50%). From the fraction of 5% ethanol–benzene, II and III were obtained. Recrystallization of the latter from ethyl acetate gave colorless crystals: mp 160–162°; $R_f$ 0.95 (A), 0.95 (B).

*Anal.* Calcd for $C_{15}H_{20}N_4O_3$: C, 59.19; H, 6.62; N, 18.41. Found: C, 59.36; H, 6.90; N, 18.35.

**7-(Tetrahydro-2-pyranyl)xanthine (VI).**—From 5 g of xanthine (0.033 mole) 4 g of crude VI was obtained in the same way. Recrystallization from acetone gave colorless crystals, which decomposed over 200°: $R_f$ 0.74 (A), 0.54 (B).

*Anal.* Calcd for $C_{10}H_{12}N_4O_3$: C, 50.8; H, 5.1; N, 23.7. Found: C, 50.22; H, 5.76; N, 23.55.

**5′-O-(tetrahydro-2-pyranyl)-2′,3′-O-isopropylideneinosine (VII).**—The reaction of 10 g of 2′,3′-O-isopropylideneinosine (0.033 mole) with 2,3-dihydro-4H-pyran resulted 7 g of crude VII which was recrystallized from ethyl acetate to give colorless crystals: mp 191–192° (dec); $R_f$, 0.89 (A), 0.89 (B).

*Anal.* Calcd for $C_{18}H_{24}N_4O_6$: C, 55.1; H, 6.12; N, 14.28. Found: C, 55.13; H, 6.35; N, 14.14.

**1-Methyl-5′-O-acetyl-2′,3′-O-isopropylideneinosine (IV).**—5′-O-Acetyl-2′,3′-O-isopropylideneinosine was added to the ether solution of diazomethane prepared from N-nitrosomethylurea. The solution stood over night at room temperature and evaporated. The residue was extracted by cyclohexane. After standing the extract precipitated crude IV: mp 85–88°; $R_f$ 0.90 (A), 0.92 (B).

*Anal.* Calcd for $C_{16}H_{20}N_4O_6$: C, 52.8; H, 5.5; N, 15.4. Found: C, 52.82; H, 6.31; N, 15.06.

**1-Methyl-5′-O-(tetrahydro-2-pyranyl)-2′,3′-O-isopropylideneinosine (VIII).**—From VII, crude VIII was obtained in the same way: mp 77°–80°; $R_f$ 0.93 (A), 0.95 (B).

*Anal.* Calcd for $C_{19}H_{26}N_4O_6$: C, 56.3; H, 6.4; N, 13.8. Found: C, 56.17; H, 7.03; N, 13.48.

## Aryliodosodifluorides

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The aryliodosodifluorides have received limited attention[1–4] as fluorinating agents, possibly because of the difficulty involved in their preparation and storage. A method is now presented which allows a simple and rapid preparation of the reagent. It involves a one-step reaction of mercuric oxide and aqueous hydrofluoric acid with the iodosodichloride in methylene chloride. The methylene chloride solution is then used directly for fluorination.

$$ArICl_2 + HgO + 2HF \longrightarrow ArIF_2 + H_2O + HgCl_2$$

**1a**, Ar = phenyl
**b**, Ar = *p*-chlorophenyl
**c**, Ar = *p*-tolyl
**e**, Ar = *p*-nitrophenyl

Previously, phenyliodosodifluoride has been prepared from iodosobenzene and hydrofluoric acid.[1] Iodosobenzene has been prepared from phenyliodosodichloride or diacetate by base hydrolysis.[5] However, iodosobenzene, upon standing, disproportionates slowly into iodobenzene and iodoxybenzene, which cannot be converted to **1a** by treatment with hydrofluoric acid.

When Bockemüller treated 1,1-diphenylethylene with **1a** he obtained a difluoro compound which he assumed to be 1,1-diphenyl-1,2-difluoroethane (**2**). The compound has now been shown by nmr studies to be 1,2-diphenyl-1,1-difluoroethane (**3**); the same difluoro derivative, **3**, is produced from 1,1-diphenylethylene and lead tetrafluoride.[6]

$Ph_2CFCH_2F$ **2** $\qquad$ $PhCF_2CH_2Ph$ **3**

Bockmüller noted that fluorination did not proceed without some added hydrogen fluoride. The methylene chloride solution of **1** generated by our procedure has enough hydrogen fluoride dissolved in it to catalyze

(1) O. Dimroth and W. Bockemüller, *Ber.*, **64B**, 516 (1931).
(2) E. Gudriniece, O. Nieland, and G. Vanags, *Zh. Obsch. Khim.*, **27**, 2737 (1957); *Chem. Abstr.*, **52**, 7177 (1958).
(3) P. G. Holton, A. D. Cross, and A. Bowers, *Steroids*, **2**, 71 (1963).
(4) B. S. Garvey, Jr., L. F. Halley, and C. F. H. Allen, *J. Am. Chem. Soc.*, **59**, 1827 (1937).
(5) (a) "Organic Syntheses," Coll. Vol. III, John Wiley and Sons, Inc., New York, N. Y., 1955, p 483; (b) *ibid.*, p 482. (c) *Org. Syn.*, **43**, 60 (1963).
(6) J. Bornstein and M. R. Borden, *Chem. Ind.* (London), 441 (1958).