Table I

	GLC-glass capillary column, R_{pim}	
	SE-30 (170 °C)	DBS (190 °C)
10 (this work)	0.862	0.903
11 (this work)	0.801	0.813
10[7]	0.862	0.903
11[7]	0.799	0.811
11[4]	0.800	0.809

1.12 g (25%) of lactone **5**: $[\alpha]^{25}_D$ −21.9° (*c* 0.19, $CHCl_3$); IR 1725 (δ-lactone) cm^{-1}; NMR (270 MHz, $CDCl_3$) 7.23, 7.70, 7.00 (4 aromatic protons), 4.19 (q, *J* = 7 Hz, H-21), 2.86 (sept, *J* = 7 Hz, H-15), 2.60 (t, 2 H, *J* = 8 Hz, H-7), 1.32 (d, C-9 methyl), 1.25 (C-4 methyl), 1.23 (d, isopropyl methyls), 0.84 ppm (C-10 methyl); mass spectrum, *m/e* 328 (M^+).

Anal. Calcd for $C_{22}H_{32}O_2$: C, 80.44; H, 9.82. Found: C, 80.09; H, 9.86.

Preparation of 6a. A solution of 2 g of **4a** in 60 mL of THF and 20 mL of H_2O was stirred with 90 mg of OsO_4 for 10 min (N_2 atmosphere). After addition of 10 g of sodium metaperiodate the mixture was stirred for 24 h, filtered, and extracted with ether. The washed and dried ether extract was evaporated and the residue chromatographed over silica gel; 10% ether–hexane eluted 1.32 g (66%) of **6a** as a gum: IR 1730–1720 (double strength), 1605 cm^{-1}; NMR 9.12 (H-21), 7.18, 7.05, and 6.92 (four aromatic protons), 3.67 (OMe), 2.87 (sept, *J* = 7 Hz, H-15), 1.27, 1.23 (d), 1.23 (d), and 1.13 ppm (methyls); mass spectrum, *m/e* 344 (M^+), 255, 187, 146, 133, 131, 123, 117, 109, 105, 101, 91, 81, 55. Because the substance underwent autooxidation to **6b** (vide infra) it was not analyzed. Decarbonylation of **6a** with tris(triphenylphosphine)rhodium(I) chloride could not be achieved in benzene solution at 80 °C or in benzonitrile at 180 °C.

Preparation of 6b. To a solution of 0.1 g of $KMnO_4$ and 10 g of $NaIO_4$ in 400 mL of H_2O and enough 3% aqueous K_2CO_3 to maintain the pH at 6 was added 1.83 g of **4a** in 300 mL of *tert*-butyl alcohol. The mixture was stirred at room temperature for 3 days, acidified with 1 N HCl, and extracted with $CHCl_3$. After being washed, dried, and evaporated, the $CHCl_3$ extract was chromatographed over silica gel. $CHCl_3$ eluted 1.66 g (86%) of **6b** as a gum: $[\alpha]^{25}_D$ 13.8° (*c* 0.087, $CHCl_3$); IR 3500–2500, 1730, 1695, 1605 cm^{-1}; NMR (270 MHz, $CDCl_3$) 7.16, 7.02, and 6.93 (four aromatic protons), 3.67 (OMe), 2.84 (sept, *J* = 7 Hz, H-15); mass spectrum, *m/e* 360 (M^+) 328, 147, 146, 134, 133, 131, 123, 121, 117, 109, 105, 96, 93, 92, 91, 81, 79, 67, 55. The elemental analysis remained somewhat unsatisfactory.

Anal. Calcd for $C_{22}H_{32}O_4$: C, 73.30; H, 8.95. Found: C, 72.29; H, 8.94.

Oxidative Decarboxylation of 6b. A. A solution of 1 g of **6b** in 50 mL of benzene (distilled over CaH_2 and $Pb(OAc)_4$) and 10 mL of pyridine (distilled over KOH and $Pb(OAc)_4$) was stirred for 20 min with 60 mg of $Cu(OAc)_2 \cdot H_2O$ in an argon atmosphere free of oxygen. $Pb(OAc)_4$ (3 g) was added and stirring was continued for 1 h. The mixture was then transferred to a quartz tube; a slow stream of oxygen-free dry argon gas was passed through the solution for 45 min after which the mixture was photolyzed in a Rayonet photochemical reactor for 6 h while a slow stream of argon was bubbled through. The mixture was diluted with 20 mL of ethylene glycol and then H_2O and extracted with ether. The washed and dried ether extract was evaporated and chromatographed over silica gel; 3% ether–hexane eluted 0.42 g (48%) of a 57:36:7 mixture of **7**, **8**, and **9** (NMR analysis) as a viscous oil: IR 1730, 1645, 1605 cm^{-1}; NMR (270 MHz, $CDCl_3$) 7.21 and 7.04 (aromatic protons), 5.27 (m, H-1 of **9**), 4.89 and 4.72 (H-20 of **8**), 3.64, 3.62, and 3.60 (OMe of **9**, **8**, and **7**, respectively), 2.85 (sept, *J* = 7 Hz, H-15), 1.73 and 170 (C-10 methyl of **9** and **8**, respectively), 1.23 (d, *J* = 7 Hz, isopropyl methyls of all three compounds), 1.19, 1.17, and 1.04 ppm (C-4 methyl of **9**, **8**, and **7**, respectively); mass spectrum, *m/e* 314, 255, 181, 147, 146, 133, 131, 121, 109, 91.

Anal. Calcd for $C_{21}H_{30}O_2$: C, 80.21; H, 9.62; mol wt 314.2245. Found: C, 79.60; H, 9.69; mol wt (mass spectrum) 314.2279.

Further elution with 5% ether–hexane furnished 0.25 g (30%) of **1** as a gum: $[\alpha]^{25}_D$ 9.5° (*c* 0.11, $CHCl_3$); IR 1770, 1603 cm^{-1}; NMR (270 MHz, $CDCl_3$) 7.24 and 7.05 (4 aromatic protons), 2.89 (sept, *J* = 7 Hz, H-15), 2.65 (m, benzylic hydrogens), 1.38 (C-10 methyl), 1.24 (d, *J* = 7 Hz, isopropyl methyls), 1.16 ppm (C-4 methyl); mass spectrum, *m/e* 300 (M^+), 256, 146, 133, 131, 123, 122, 121, 117, 111, 110, 109, 91, 81, 67. In a second experiment, 1.12 g of **7** furnished 0.42 g (47%) of the mixture of **7**, **8**, and **9** and 0.27 g (29%) of **10**. The synthetic material was identical in all respects (NMR, TLC, GLC, IR) with an authentic sample supplied by Drs. J. W. Rowe and A. H. Conner.

Preparation of 10 and 11. Hydrogenation of 0.3 g of the mixture of **7**, **8**, and **9** in 25 mL of EtOAc over 30 mg of PtO_2 at 50 psi and room temperature for 24 h, filtration, evaporation, and high-performance LC (4 ft × $^3/_8$ in. Porasil B, 1% ether–heptane) gave 0.14 g of a mixture of **10** and **11** as a gum which exhibited two peaks on GLC (Apiezon M): IR 1730, 1603, 1587, 1487 cm^{-1}; NMR (270 MHz, $CDCl_3$) 7.19 and 7.00 (aromatic protons), 3.67 and 3.65 (OMe of **10** and **11**, respectively), 2.85 (sept, *J* = 7 Hz, H-15), 2.60 (t) and 2.59 (t) (*J* = 9 Hz, benzylic protons of **11** and **10**, respectively), 1.25 (d) and 1.23 (d) (*J* = 7 Hz, isopropyl methyls of **10** and **11**), 1.15 and 1.13 (C-4 methyls of **10** and **11**), 1.01 (d) and 0.91 (d) (*J* = 7 Hz, C-10 methyls of **11** and **10**); mass spectrum, *m/e* 316, 284, 192, 187, 147, 134, 133, 131, 123, 117, 109, 105, 101, 95, 92, 91, 81, 69, 67, 55. Further elution with the same solvent system furnished 0.09 g of a 2:1 mixture of **8** and **9** (NMR analysis). Further hydrogenation of 70 mg of this mixture (PtO_2, H_2 at 50 psi, 24 h), followed by high-performance LC, resulted in formation of 4 mg of a mixture of **10** and **11** and recovery of 65 mg of a mixture of **8** and **9** in which the proportion of **8** had decreased slightly. Evidently reduction of **8** under these conditions proceeds only very slowly and reduction of **9** not at all. This eliminates the possibility that hydrogenation of **7** might actually have given rise to a mixture of **10** and **12** by prior isomerization to **9** and subsequent reduction.

A direct GLC comparison of the synthetic mixture of **10** and **11** with samples of the methyl secodehydroabietates derived from tall oil[4] and thermal isomerization of methyl pimarate[7] was carried out by Dr. Duane Zinkel at the Forest Products Laboratory, Madison, WI. The results shown in Table I established their identity.

Acknowledgment. We are indebted to Dr. Duane Zinkel for carrying out the GLC comparison of **10** and **11** with authentic material.

Registry No. **1**, 57119-17-2; **2a**, 79-54-9; **3a**, 6512-55-6; **3b**, 75399-78-9; **4a**, 75399-79-0; **4b**, 75399-80-3; **5**, 75399-81-4; **6a**, 75399-82-5; **6b**, 75399-83-6; **7**, 75399-84-7; **8**, 75399-85-8; **9**, 75399-86-9; **10**, 75443-46-8; **11**, 19556-81-1; ethyl propiolate, 623-47-2.

Specific Dealkylation of 3-Benzyladenines in the Presence of 9-Benzyladenines

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The decomposition of onium ions, e.g., oxonium, sulfonium, and ammonium ions, constitutes an important class of carbenium ion generating reactions[1] (Scheme I). Ammonium salts derived from *N*-heterocycles have been employed as sources of carbenes,[2] nitrenes,[3] and oxenium ions,[4] but we are unaware of the formation of carbenium ions from these starting materials.[19]

We now report a specific dealkylation of 3-benzyladenines that proceeds via benzyl carbenium ion formation and the successful application of this reaction to the quantitative removal of the 3-isomer from adenine al-

† Deceased.

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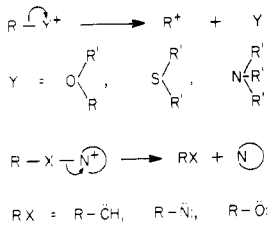
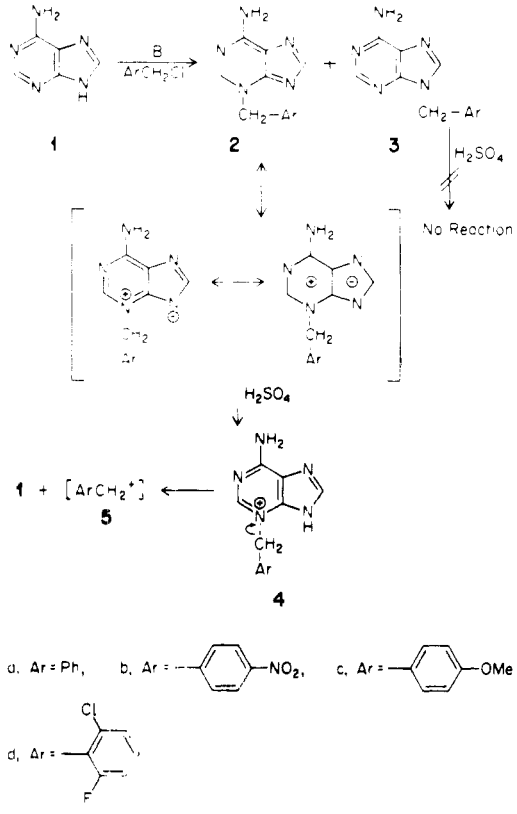
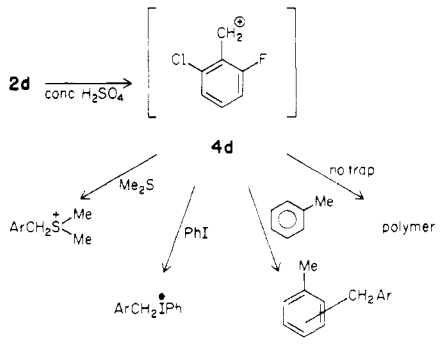
Scheme I



Scheme II



Scheme III



kylation mixtures. It has been observed by ^{1}H NMR that 3-benzyladenine (**2a**) in 96% sulfuric acid at 50 °C is rapidly decomposed to adenine (**1**) and a polymer[5] derived from the benzyl carbenium ion (**5a**). Under the same conditions, 9-benzyladenine (**3a**) remains unchanged even after extended reaction periods. The intermediacy of a benzyl carbenium ion is indicated by the failure of 3-(*p*-nitrobenzyl)adenine (**2b**) to enter the reaction coupled with the observation that 3-(*p*-methoxybenzyl)adenine (**2c**) is rapidly degraded to (**1**) at 20 °C even in 90% sulfuric acid. In every case the corresponding 9-isomer (**3a–c**) remained unchanged.

These transformations parallel the relative thermodynamic stabilities of the isomeric benzylated adenines[7] which decrease in the order of 9- > 7- > 3-. The purine rings in the 3-isomer **2** are more polarized than those in the 9-isomer **3**; i.e., the positive charge in the pyrimidine moiety and the negative charge in the imidazole ring of **2** are larger than those of the 9-isomers.[8] Protonation of the 3-isomer would be expected to produce the adeninium salt **4a** followed by cleavage to **5a** and adenine (Scheme II).

This specific debenzylation was applied to the quantitative purification of the important coccidiostat arprinocid,[9] 9-(2-chloro-6-fluorobenzyl)adenine,[10] **3d**. Since the 3-isomer **2d** gives a weak positive Ames test, its virtually complete removal to levels less than 50 ppm was mandatory. Due to formation of a solid solution[14] of **2d** and **3d**, conventional purification[15] methods as well as selective acid extraction (pK_a **3d**, 4.0; pK_a **2d**, 5.6) failed to reduce the unwanted 3-isomer to less than 5000–10000 ppm.

By taking advantage of its inherent lower thermodynamic stability, it was possible to reduce levels of **2d** to less than 10 ppm via the specific acid degradation discussed above. As expected, **2d** was rapidly decomposed to adenine and a polymer derived from the 2-chloro-6-fluorobenzyl carbenium ion whereas under the same conditions **3d** remained unchanged. Preliminary kinetic data were obtained by ^{19}F NMR studies monitoring the disappearance of **2d**. The half-life of **2d** in 96% and 90% sulfuric acid at 25 °C was found to be 5 and 24 min, respectively.

On a preparative scale an 85/15 mixture of **3d** and **2d**[12] on treatment with 96% sulfuric acid at 60 °C resulted in a 96% recovery of **3d** containing less than 10 ppm of **2d**. It proved difficult, however, to completely remove the polymeric byproduct. This problem was overcome by carrying out the reaction in presence of suitable carbenium ion traps, namely, dimethyl sulfide, iodobenzene, and toluene, which lead, respectively, to quantitative formation

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(14) The 3-isomer **2d** level reaches the range of 0.05–0.1% even though the liquid phase is not saturated in **2d**. This is due to the pronounced tendency toward solid solution formation by **2d** and **3d**. We thank Dr. J. A. McCauley of our analytical chemistry department for the studies related to solid solutions.
(15) It is difficult to obtain pure 9-substituted adenine without chromatographic separation, see: Yamauchi, K; Tanabe, T.; Kinoshita, M. *J. Org. Chem.* **1976**, *41*, 3691.

of sulfonium salts, iodonium salts, and an ortho–meta–para mixture of 2-chloro-6-fluoro benzylated toluenes (Scheme III).

The reaction of the 85/15 mixture of **3d** and **2d** in 96% sulfuric acid in the presence of toluene gave pure **3d** containing less than 10 ppm of **2d** in 98% recovery. Recently, specific hydrogenation of 3-benzyladenine **2a** to adenine and toluene was reported.[16] This method was not applicable in our case, however, due to the presence of the chlorine atom in **2d** which was partially removed during the hydrogenation.

In summary, the specific dealkylation of 3-benzyladenines in concentrated sulfuric acid to form adenine and benzyl carbenium ions was observed. The carbenium ion was trapped via transalkylation with toluene in order to prevent polymer formation. Accordingly, this observation was successfully applied to the purification of crude alkylation mixtures to obtain pure 9-benzyladenines.

Experimental Section

General Methods. ^{1}H NMR spectra was recorded at 60 MHz with a Hitachi Perkin-Elmer R-24A instrument and at 100 MHz with a Varian Associates XL-100 instrument. Me_4Si was used as an internal standard. ^{19}F NMR spectra were obtained with a Varian XL-100 instrument at 94.1 MHz. The product ratio of **2d** and **3d** was determined by high-performance LC[17] using a 5-μm porous silica column (Du Pont, Zorbax-SIL) eluted with $CHCl_3$/MeOH (95/5) and detected at 254 nm. The 3-isomer analysis at the parts per million level (less than 10 ppm) was achieved by high-performance LC using a reverse-phase column.[17]

Alkylation of Sodium Adeninate. The following is a typical preparation. Into a 250-mL three-necked flask fitted with a mechanical stirrer, thermometer, and pressure-equalized dropping funnel, connected to a Firestone valve (Ace Glass Co., Vineland, NJ, Catalog No. 8766-12), were charged sodium adeninate (7.85 g, 50 mmol) and 100 mL of sieve-dried acetone containing 1.25 g of Aliquat 336 (a mixture of tetraalkylammonium salts in which the alkyl groups are primarily caprylyl, manufactured by General Mills).

To this suspension was added dropwise over a 10-min period a solution of α,2-dichloro-6-fluorotoluene (9.8 g, 50 mmol) in 10 mL of acetone at room temperature and the resulting mixture was boiled under reflux for 6 h. The reaction mixture was cooled to room temperature, and the solids were collected by filtration, washed with acetone (2 × 15 mL) and then stirred with 50 mL of 0.1 N sodium hydroxide for 15 min to remove unreacted sodium adeninate and NaCl. The solid was collected by filtration, washed with water (2 × 20 mL), and dried in vacuo to give 13.1 g (yield 95%) of a mixture of **2d** and **3d**. This mixture was recrystallized three times from acetic acid–water, affording **3d** (high-performance LC, wt % of **3d** = 99.6%; **2d** = 0.3%). A sample of **2d** was obtained from the mother liquor (high-performance LC, wt % of **3d** = 0.6%; **2d** = 99.4%). Similarly, the alkylation of 1 with benzyl, *p*-nitrobenzyl, and *p*-methoxybenzyl chloride gave the crude alkylation mixtures **2a**/**3a**, **2b**/**3b**, and **2c**/**3c**, respectively.[12]

Dealkylation of 2d in the Presence of Toluene. To a vigorously stirred suspension of a crude alkylation mixture consisting of an 85/15 ratio of **2d** and **3d** (50 g, 0.2 mole) in 100 mL of toluene was added dropwise 96% sulfuric acid (100 mL) with ice–water cooling as required to maintain a temperature of 50–60 °C. The mixture was heated with stirring at 60 °C for 8 h, cooled to room temperature, and poured into 300 mL of ice–water. The mixture was transferred to a steam-jacketed separatory funnel and heated to 90 °C in order to redissolve the precipitate. The aqueous layer (about 400 mL) was separated at 90 °C and washed with 50 mL of hot toluene to remove the transalkylation product. The aqueous layer was made basic (pH 10) at 50 °C by addition of concentrated ammonium hydroxide. The precipitated, colorless solid was collected and washed with boiling water (3 × 100 mL) and 50% methanol (2 × 100 mL). The yield of **3d** was 47.8 g (99%): mp 247–248 °C; high-performance LC wt % of **3d** = 100.01%; 3-isomer **2d**, none detectable (<10 ppm). Anal. Calcd for $C_{12}H_9N_5ClF$: C, 51.90; H, 3.27; N, 25.22; Cl, 12.77. Found: C, 51.67; H, 3.04; N, 25.29; Cl, 12.81.

The toluene layer was concentrated in vacuo, and the residue was purified by vacuum distillation to give 6.04 g (95.3%) of an *o*-, *m*-, and *p*-(2-chloro-6-fluorobenzyl)toluene mixture: bp 120–123 °C (0.05 mmHg); mass spectrum, *m/e* 234, 236; ^{1}H NMR ($CDCl_3$) δ 2.22 and 2.37 (total 3, CH_3), 4.1 (m, 2, CH_2), 6.9 (m, 7, aromatic protons). The ortho–meta–para ratio was found to be 2:1:3.5, obtained by ^{13}C NMR.[18] Anal. Calcd for $C_{14}H_{12}FCl$: C, 71.64; H, 5.16. Found: C, 71.48; H, 5.09.

Dealkylation of 2d in the Presence of Dimethyl Sulfide. This reaction was carried out the same as above, using dimethyl sulfide in place of toluene. The reaction mixture was sampled by aliquot and examined by ^{1}H NMR after dilution with CD_3COOD. The new doublet ($^4J_{HF}$ = 1.7 Hz) was observed at 1.2 ppm higher field than the methylene peak in **2d** accompanied by a singlet at 1.0 ppm lower field relative to dimethyl sulfide. This ^{1}H NMR observation supports the structure of 2-chloro-6-fluorobenzyldimethylsulfonium salt. After the usual workup a 98% yield of **3d** was obtained. High-performance LC wt % of **3d** = 99.6%. There was less than 10 ppm of **2d** observed by high-performance LC. Dealkylation of **2d** in the presence of iodobenzene was carried out the same as above and **3d** was obtained (86% yield) which showed a satisfactory high-performance LC analysis of the 3-isomer level.

Acknowledgment. We thank Mr. A. M. DeMarco for his technical assistance.

Registry No. 1, 73-24-5; **2a**, 7280-81-1; **2b**, 75347-16-9; **2c**, 75347-17-0; **2d**, 68220-23-5; **3a**, 4261-14-7; **3b**, 5134-49-6; **3c**, 56046-26-5; **3d**, 55779-18-5; α,2-dichloro-6-fluorotoluene, 55117-15-2; benzyl chloride, 100-44-7; *p*-nitrobenzyl chloride, 100-14-1; *p*-methoxybenzyl chloride, 824-94-2; *o*-(2-chloro-6-fluorobenzyl)toluene, 75347-18-1; *m*-(2-chloro-6-fluorobenzyl)toluene, 75347-19-2; *p*-(2-chloro-6-fluorobenzyl)toluene, 75347-20-5.

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(17) We thank Dr. W. Caldwell and Miss B. C. Rettberg for high-performance LC assay for **2d** and **3d**. Detailed assay work will be published elsewhere.

(18) We thank Mr. R. A. Reamer for the ^{13}C NMR studies.

(19) Formation of oxocarbenium ions by the solvolytic decomposition of nucleoside analogues has recently been suggested; see: Lönnberg, H.; Käppi, R. *Tetrahedron* **1980**, *36*, 913 and references therein.

Syntheses and Characterization of Some Tetra- and Pentaazaindene *N*-Oxides

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In connection with our work on 1,2,4-triazines and their *N*-oxides,[1,2] it was of interest to examine the effect that a 1-oxide in 1,2,4-triazines has upon the mode of cyclization of an appropriate 3-substituted-1,2,4-triazine 1-oxide. To this end we prepared 3-hydrazino-1,2,4-triazine 1-oxide (**1**) and treated it with the one-carbon cyclizing agent **2**, so

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