5,6,7,8,9,10,11,12,12,14-Decahydro-3-hydroxy-1-methylcyclododeca[c]pyridine-4-carbonitrile (VII).—2-Acetylcyclododecanone was treated as described above except the reaction time was 72 hr. VII, mp 281.5–282° dec, was obtained in 49.6% yield: uv max (95% EtOH) 217 mμ (log ε 4.29), 239 (infl, 3.83), 221 (4.29), 249 (3.59), 341 (4.10); ir (KBr) 3311, 3156 (NH), 2237 (CN), 1631, 1534 (C=O), 829, 778, 769 cm^{-1} (cycloalkyl).

Anal. Calcd for $C_{17}H_{24}N_2O$: C, 74.96; H 8.88; N, 10.29. Found: C, 75.04; H, 8.70; N, 10.27.

6,7,8,9-Tetrahydro-3-hydroxy-1-methyl-5H-cyclohepta[c]pyridine (VIII).—VIII was prepared from V according to the procedure of Prelog and Metzler.[10] Recrystallization from dilute ethanol gave VIII: mp 256.5–257°, in 71% yield; uv max (95% EtOH) 201 mμ (log ε 4.19), 237 (infl, 3.79), 244 (3.72), 305 (3.88); ir (KBr) 3367 (NH), 1650, 1541 (C=O), 842, 759 cm^{-1} (cycloalkyl).

Anal. Calcd for $C_{11}H_{15}NO$: C, 74.54; H, 8.53; N, 7.90. Found: C, 74.71; H, 8.56; N, 7.95.

Registry No.—I, 107-91-5; II, 21297-71-2; III, 21298-56-6; IV, 21298-57-7; V, 21298-58-8; VI, 21298-59-9; VII, 21298-60-2; VIII, 21298-61-3.

Acknowledgment.—We express our thanks to Kay-Fries Chemicals, Inc., for generous samples of malononitrile and malonoamide.

The Reaction of 1-Trimethylsilyl-1,4-dihydropyridine with Ethyl Azidoformate and *p*-Tosyl Isocyanate[1]

EMIL J. MORICONI AND ROBERT E. MISNER[2]

Department of Chemistry, Fordham University, New York, New York 10458

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The recently reported synthesis of 1-trimethylsilyl-1,4-dihydropyridine (**1**) and its base-catalyzed methanolysis product 1,4-dihydropyridine (**2**)[3] should have normally spurred an intense study of the chemistry of these very reactive substrates.[4] To date, we have not seen any study reported.[6]

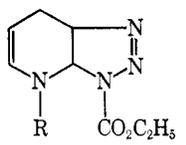
In this Note we report on two reactions of **1** with electrophiles to give ultimately substitution products at different sites. Thus, treatment of **1** with ethyl azidoformate followed by methanolysis led to a 40–55% yield of the tautomeric mixture **3a** ⇌ **3b**.[7] In the infrared the mixture displayed both free (5.79 μ) and bonded (5.90 μ) carbonyl absorptions.

Treatment of 1-phenyl-1,4-dihydropyridine (**6**) with ethyl azidoformate led only to the single product, 1-phenyl-2-carbethoxyimino-1, 2, 3, 4-tetrahydropyridine (**7**) in 35% yield.[8] Adduct **7** is incapable of tautomerism; hence the infrared showed only a single carbonyl absorption at 5.93 μ.

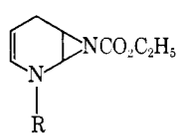
Hydrogenation of **3a** ⇌ **3b** led expectedly to the tautomeric mixture **4a**[9] ⇌ **4b** ($\lambda_{C=O}^{KBr}$ 5.78 and 5.98 μ), while dehydrogenation of **3a** ⇌ **3b** over Pd–C afforded the known ethyl 2-pyridinecarbamate (**5**) (53%)[10] prepared independently from ethyl chloroformate and 2-aminopyridine.[10b]

Treatment of **1** with *p*-tosyl isocyanate afforded N-*p*-tosyl-1-trimethylsilyl-1,4-dihydronicotinamide (**8**), in addition to large amounts of an insoluble, unidentified material (Scheme I).

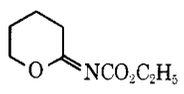
Generally, dihydropyridines are obtainable from pyridine quaternary salts *via* sodium amalgam or borohydride reductions.[5,11,12] Thus, Saunders and Gold[5] obtained 1-phenyl-1,2-dihydropyridine (60%) on sodium or potassium borohydride reduction of 1-phenylpyridinium chloride (**12**), and the 1,4 isomer (**6**, 70%) using sodium amalgam.[13] In an effort to prepare 1-benzyl-1,4-dihydropyridine (**11**) as a substrate for reactions previously discussed, the benzylpyridinium cation (**9**) was reduced with sodium amalgam. The only product isolated was 1,1′-dibenzyltetrahydro-4,4′-bipyridyl (**10**),[14] further hydrogenation of which led to the known 1,1′-dibenzyl-4,4′-bipiperidyl.[15] A repetition of the Saunders and Gold effort[5] using Karrer's work-up procedure[13] expectedly led to **6**, in addition to 1,1′-diphenyltetrahydro-4,4′-bipyridyl (**13**, 7%). Sodium borohydride reduction of **9** gave the known 1-benzyl-1,2,5,6-tetrahydropyridine (**14**)[16] and smaller amounts of 1-benzylpiperidine (**15**). Catalytic reduction of **14** afforded **15**.



I



II



III

(1) This research was supported by the Department of the Army, U. S. Army Research and Development Command Office, Office of the Surgeon General, under Contract DA-49-193-MD-2992, and by Public Health Service Research Grant 5 R01 AI-08063-01 from the National Institute of Allergy and Infectious Diseases. This is Contribution No. 533 to the Army Research Program on Malaria.

(2) Abstract from the Ph.D. Thesis of R. E. Misner.

(3) N. C. Cook and J. E. Lyons, *J. Amer. Chem. Soc.*, **88**, 3396 (1966); **87**, 3283 (1965).

(4) For example, these planar 6 π electron heterocycles contain the elements of a cross-conjugated dienamine, and both they and their salts should be responsive to electrophiles and nucleophiles, respectively. Alkylation, however, of 1-phenyl-1,4-dihydropyridine with methyl iodide led only to 1-phenylpyridinium iodide.[5]

(5) M. Saunders and E. H. Gold, *J. Org. Chem.*, **27**, 1439 (1962).

(6) We perhaps can understand why: the reaction of **1** with Br_2, 70% perchloric acid, methyl iodide, ethyl acrylate, chlorosulfonyl isocyanate, carbenes, and Simmons–Smith reagent led to no isolable products.

(7) *Cf.* similar tautomerism observed with 2-amino- and 2-alkylaminotetrahydropyridines [T. B. Grave, *J. Amer. Chem. Soc.*, **46**, 1460 (1924); M. Freifelder, R. W. Mattoon, and Y. H. Ng, *J. Org. Chem.*, **29**, 3730 (1964)].

(8) Mechanistically, products **3** and **7** could be accounted for *via* collapse of such initial adducts as the triazoline I and aziridine II. No such intermediates were experimentally observed, however.

(9) The oxygen analog of **4a**, III, was spectrally inferred as the primary product of the thermal reaction between dihydropyran and ethyl azidoformate. The nmr spectrum of III was quite similar to that of **4a** ⇌ 4b [I. Brown and O. E. Edwards, *Can. J. Chem.*, **43**, 1266 (1965)].

(10) (a) R. L. Shriner and R. G. Child, *J. Amer. Chem. Soc.*, **74**, 549 (1952); (b) A. R. Katritzky, *J. Chem. Soc.*, 2063 (1956).

(11) R. A. Barnes, "Pyridine and its Derivatives," Part I, Vol. 14, E. Klingsberg, Ed., Interscience Publishers, Inc., New York, N. Y., 1960, p 77.

(12) F. Liberatore, V. Carelli, and M. Cardellini, *Tetrahedron Lett.*, 4735 (1968).

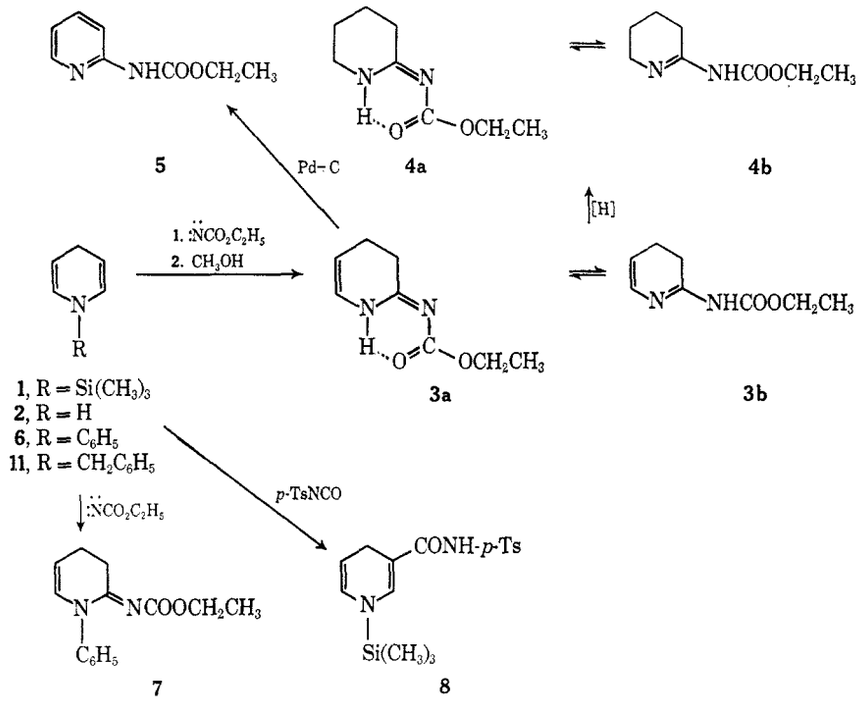
(13) P. Karrer, G. Schwarzenbach, and G. E. Utzinger, *Helv. Chim. Acta*, **20**, 72 (1937).

(14) A. W. Hoffman, *Ber.*, **14**, 1497 (1881); B. Emmert, *ibid.*, **42**, 1997 (1909).

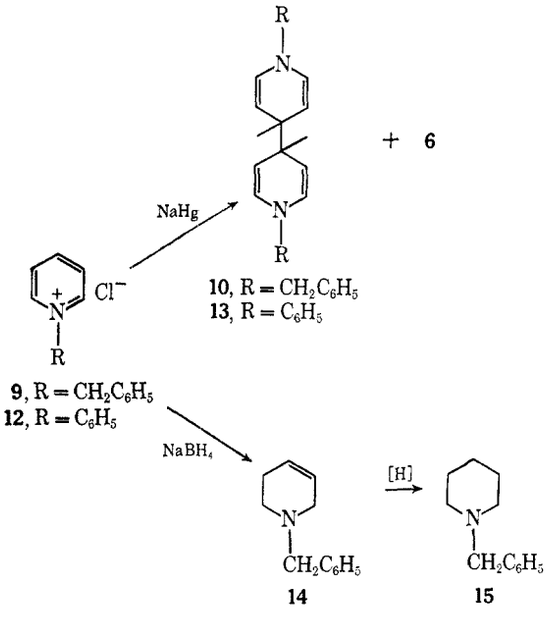
(15) H. Ochai and K. Kataoka, *J. Pharm. Soc. Jap.*, **62**, 241 (1942).

(16) A. N. Kost, L. G. Yudin, and A. E. Shipov, *Chem. Abstr.*, **52**, 9115*b* (1948); P. S. Anderson, Ph.D. Thesis, University of New Hampshire, Durham, N. H., 1963; V. Petrov and O. Stephenson, *J. Pharm. Pharmacol.*, **14**, 306 (1962).

SCHEME I



SCHEME II



Experimental Section[17]

Reaction of 1-Trimethylsilyl-1,4-dihydropyridine (1) with Ethyl Azidoformate.—To a solution of 15.3 g (0.01 mol) of **1** dissolved in 50 ml of degassed benzene was added, at once, 23.0 g (0.02 mol) of ethyl azidoformate. The solution was stirred (with initial cooling) for 4 hr at room temperature under an atmosphere of nitrogen. The solvent was removed *in vacuo* and the residue was distilled at 93–97° and 0.2 mm to yield 16.0 g of a yellow oil which was dissolved in 30.0 ml of methanol. The solution was stirred (with initial cooling) at room temperature for 3.5 hr under nitrogen. The solvent was removed *in vacuo* and the residue was crystallized (Darco) from pentane to yield 9.2 g (55%) of the tautomeric mixture **2-carbethoxyimino-1,2,3,4-tetrahydropyridine** (**3a**) and **2-carbethoxyamino-3,4-dihydropyridine** (**3b**), vpc of which showed two poorly resolved peaks with almost identical retention times (column temperature 150°; He flow 20 cc min^{-1}).[18] The mixture was obtained as white needles: mp 75–76°; ir (KBr) 3.18 (**NH**), 5.79 and 5.90 (C=O), and 6.01 μ (C=N); uv max (95% EtOH) 286 mμ (ϵ 17,800); nmr (DMSO-d_6) δ 10.0 (s, 1, **NH**), 6.40–6.13 (m, 1, J = 7.5 Hz, C-6 hydrogen), 5.35–5.00 (m, 1, J = 7.5 Hz, C-5 vinyl), 4.02 (q, 2, J = 7.2 Hz, CH_2CH_3), 2.75–2.00 (m, 2, C-3 methylene), and 1.35–1.05 (t, 3, J = 7.2 Hz, CH_3); mass spectrum (70 eV) m/e 168.

Anal. Calcd for $C_8H_{12}N_2O_2$: C, 57.13; H, 7.18; N, 16.65; mol wt, 168. Found: C, 57.36; H, 7.13; N, 16.46; mol wt, 156.

Hydrogenation of 3a ⇌ 3b.—A solution of 9.2 g (0.055 mol) of **3a ⇌ 3b** in 60 ml of ethyl acetate was hydrogenated (10% Pd–C, 3 atm) in a Parr apparatus for 4 hr. The mixture was then filtered and the filtrate evaporated *in vacuo*; the material crystallized on cooling. The solid was taken up in pentane (Darco), filtered, cooled, and refiltered to yield 6.7 g (72%) of white needles. Two more recrystallizations from pentane gave the mixture **2-carbethoxyiminopiperidine** (**4a**) and **2-carbethoxyamino-3,4,5,6-tetrahydropyridine** (**4b**), vpc of which again showed two poorly resolved peaks (column temperature 150°; He flow 20 cc min^{-1}): mp 76–77°; ir (KBr) 3.12 (NH), 5.78 and 5.98 (C=O), and 6.01 μ (C=N); uv max (95% EtOH) 240 mμ (ϵ 12,000); nmr (DMSO-d_6) δ 9.71 (s, 1, **NH**), 4.18–3.78 (q, 2, J = 7 Hz, CH_2CH_3), 3.50–3.10 (m, 2, C-6 methylene), 2.60–2.20 (m, 2, C-3 methylene), 1.68–1.50 (m, 4, C-4, -5 methylenes), and 1.30–1.02 (t, 3, J = 7 Hz, CH_3).

Anal. Calcd for $C_8H_{14}N_2O_2$: C, 56.45; H, 8.29; N, 16.46. Found: C, 56.61; H, 8.28; N, 16.68.

Dehydrogenation of 3a ⇌ 3b.—A mixture of 1.0 g (0.006 mol) of **3a ⇌ 3b** and 0.25 g of 5% Pd–C in 40 ml of 95% ethanol was refluxed for 3.5 hr. The solution was then filtered while hot and the solvent was removed *in vacuo* to yield a semisolid. This material was taken up in pentane and deposited on a 2.5 × 25 cm silica gel column. Elution with 200 ml of ethyl ether gave

(17) Melting points are corrected; boiling points are uncorrected. Infrared spectra were obtained on a Perkin-Elmer 337 grating spectrophotometer. Microanalyses and molecular weights (isothermal distillation) were determined by Schwartzkopf Microanalytical Laboratory. Ultraviolet spectra were recorded on a Cary Model 15 dual beam recording spectrophotometer. Nmr spectra were determined at 60 Mc with a Varian Model A-60 spectrometer; chemical shifts are expressed as δ values in parts per million downfield from tetramethylsilane. Mass spectra were obtained on a Perkin-Elmer Hitachi Mass spectrometer. Gas chromatographic analyses were run on a Perkin-Elmer 880 instrument using a column packed with 10% SE-30 Chromosorb W (80–100 mesh).

(18) Similarly, irradiation of a mixture of **1** (5.0 g, 0.033 mol) and ethyl azidoformate (8.05 g, 0.066 mol) in 90 ml of degassed hexane at 2537 Å (Hanovia 10-W low pressure Hg vapor lamp) for 48 hr gave 2.04 g (40%) of the tautomeric mixture **3a ⇌ 3b**.

0.26 g (53%) of **ethyl 2-pyridinecarbamate (5)**. Two recrystallizations from ethanol–water led to a white solid, mp 100.5–102.5° (lit.[10b] mp 102–103°), identical in all respects with the compound prepared from the condensation of ethyl chloroformate and 2-aminopyridine:[10b] ir (KBr) 3.17 (NH) and 5.82 μ (C=O); uv max (95% EtOH) 229 mμ (ε 17,400) and 287 (6650); nmr DMSO-d_6) δ 10.3 (s, 1, NH), 8.35–8.28 (m, 1, C-6 hydrogen), 7.95–7.85 (m, 2, aromatic), 7.30–7.00 (m, 1, aromatic), 4.44–4.05 (q, 2, J = 7 Hz, CH_2CH_3), and 1.40–1.16 (t, 3, J = 7 Hz, CH_3).

Anal. Calcd for $C_8H_{10}N_2O_2$: C, 57.82; H, 6.06; N, 16.85. Found: C, 57.66; H, 6.12; N, 17.00.

Photolysis of Ethyl Azidoformate in the Presence of 1-Phenyl-1,4-Dihydropyridine (6).—1-Phenyl-1,4-dihydropyridine[5] (**6**, 1.28 g, 0.0081 mol) in 90 ml of degassed hexane and 1.76 g (0.015 mol) of ethyl azidoformate under nitrogen were irradiated for 21 hr at 2537 Å with a Hanovia 10-W low-pressure mercury vapor lamp. The solution was filtered and the filtrate was evaporated *in vacuo* to leave a crystalline residue. The residue was dissolved in ethyl ether (Darco), filtered, and pentane was added to the cloud point. Upon cooling there was obtained 0.70 g (36%) of **1-phenyl-2-carbethoxyimino-1,2,3,4-tetrahydropyridine (7)** as white needles (from ether–pentane): mp 95–96°; ir (KBr) 5.93 (C=O) and 6.18 μ (C=N); uv max (95% EtOH) 245 mμ (ε 9500) and 284 (8900); nmr (DMSO-d_6) δ 7.50–7.06 (m, 5, aromatic), 6.40–6.16 (m, 1, J = 7 Hz, NCH=), 5.40–5.10 (m, 1, J = 7.5 Hz, =CH), 4.18–3.84 (q, 2, J = 6.9 Hz, $-CH_2CH_3$), 2.94–2.20 (m, 4, C-3, -4 methylenes), and 1.16 (t, 3, J = 6.9 Hz, CH_3); mass spectrum (70 eV) m/e 244.

Anal. Calcd for $C_{14}H_{16}N_2O_2$: C, 68.83; H, 6.60; N, 11.47; mol wt, 244. Found: C, 68.84; H, 6.89; N, 11.80; mol wt, 232.

Reaction of 1 with *p*-Toluenesulfonyl Isocyanate.—A solution of 1.78 g (0.012 mol) of **1** in 200 ml of degassed anhydrous ethyl ether was brought to 0° under nitrogen. To this mixture was added slowly (45 min) 4.64 g (0.024 mol) of *p*-toluenesulfonyl isocyanate in 50 ml of anhydrous ethyl ether (degassed). Shortly after the start of the addition of the isocyanate, a solid appeared. The mixture was stirred at 0° for 3 hr, after which an unidentified solid (2.1 g) was filtered. Pentane was added to the filtrate and an additional 1.5 g of the same material was filtered. The filtrate was cooled to −20° and then filtered to yield 1.0 g of **N-*p*-toluenesulfonyl-1-trimethylsilyl-1,4-dihydronicotinamide (8)** (22%). One recrystallization (Darco) from anhydrous ethyl ether–pentane gave **8** as yellow plates: mp 123.5–125°; ir (KBr) 3.12 μ (NH), 6.00 (C=O), 7.52, 8.63 (SO_2), 7.95 (Si–CH_3) and 11.80 (Si–C); uv max (95% EtOH) 224 mμ (ε 29,500) and 355 (12,500); nmr ($CDCl_3$) δ 8.39–8.32 (m, 1, NH), 8.09–7.25 (A_2B_2 pattern, 4, J = 8.7 Hz, aromatic), 7.17 (d, 1, J = 2 Hz, C-2 vinyl), 5.90–5.57 (m, 1, J = 2 Hz, C-6 vinyl), 4.93–4.63 (m, 1, J = 3 Hz, C-5 vinyl), 3.12–2.98 (d, 2, J = 3 Hz, CH_2), 2.44 (s, 3, *p*-CH_3), and 0.18 (s, 9, CH_3).

Anal. Calcd for $C_{16}H_{22}N_2O_3SSi$: C, 54.83; H, 6.33; N, 7.99; mol wt, 350. Found: C, 54.82; H, 6.24; N, 8.26; mol wt, 350.

Na–Hg Reduction of 1-Benzylpyridinium Chloride (9).—To a solution of 11.4 g (0.055 mole) of **9**[19] in 1 l. of degassed water was added 120 g (0.026 mol) of 5% sodium amalgam. The mixture was stirred for 16 hr at ambient temperature under nitrogen. The product separated as a gray solid on continued stirring and was filtered. The residue was dissolved in degassed acetone and the solution was brought to the cloud point with water. Cooling and filtration led to 6.0 g (63%) of **1,1′-dibenzyltetrahydro-4,4′-bipyridyl (10)** as chunky white cubes: mp 86–88° [from acetone–water (Darco)] (lit.[14] no melting point reported); uv max (95% EtOH) 228 mμ (ε 12,500) and 285 (3640); nmr ($CDCl_3$) δ 7.26 (s, 10, aromatic), 5.99–5.83 (d, 4, J = 8 Hz, C-2, -2′, -6, -6′ vinyl), 4.50–4.28 (m, 4, C-3, -3′, -5, -5′ vinyl), 4.13 (s, 4, $CH_2C_6H_5$), and 3.00 (m, 2, C-4, -4′ methine).

Anal. Calcd for $C_{24}H_{24}N_2$: C, 84.67; H, 7.11; mol wt, 340. Found: C, 84.68; H, 7.38; mol wt, 362.

Hydrogenation (PtO_2) of **10** in ethyl acetate at 1 atm for 4.5 hr led to **1,1′-dibenzyl-4,4′-bipiperidyl**, mp 133–134° (from anhydrous ethyl ether), lit.[15] mp 133°.

$NaBH_4$ Reduction of 9.—To a solution of 13.8 g (0.25 mol) of KOH in 1 l. of degassed water was added 25.2 g (0.12 mol) of **9**. To this mixture was added 1.67 g (0.044 mol) of $NaBH_4$ in 50 ml of water and the whole was stirred for 16 hr at room temperature under nitrogen. The aqueous solution was extracted with eight 100-ml portions of ethyl ether. The ether extracts were combined, dried ($MgSO_4$), and filtered, and the solvent was removed *in vacuo*. The residue was distilled at 78–84° (0.25 mm) to yield 10.7 g (51%) of a colorless (two-component by vpc) liquid. Chromatography over a 20 ft × 0.375 in. 20% Silicone rubber on 60–80 Chromosorb W column (column temperature 150°, He flow 30 cc min^{-1}) separated the larger component, bp 69–70° (0.2 mm), which was identical with the known **1-benzyl-1,2,5,6-tetrahydropyridine (14)** [lit.[16] bp 68° (0.1 mm); 96° (5 mm)]; uv max (95% EtOH) 251 mμ (ε 300), 257 (335), and 263 (332); nmr ($CDCl_3$) δ 7.23 (s, 5, aromatic), 5.73–5.50 (m, 2, CH), 3.54 (s, 2, $-CH_2C_6H_5$), 3.03–2.80 (m, 2, C-2 methylene), 2.68–2.41 (m, 2, C-6 methylene), and 2.36–2.00 (m, 2, C-5 methylene).

Anal. Calcd for $C_{12}H_{15}N$: C, 83.18; H, 8.57. Found: C, 83.01; H, 8.57.

Hydrogenation (5% Pd–C) of the initial mixture (3 ml, 0.016 mol) obtained from the $NaBH_4$ reduction of **9** in 50 ml of absolute ethanol at 1 atm gave 1.6 g (58%) of **1-benzylpiperidine (15)**, bp 120–122 (13 mm) [lit.[16] bp 110° (6 mm)]; nmr ($CDCl_3$) δ 7.29 (s, 5, aromatic), 3.41 (s, 2, $-CH_2C_6H_5$), 2.50–2.24 (m, 4, C-2, -6 methylenes), and 1.66–1.40 (m, 6, C-3, -4, -5 methylenes).

Na–Hg Reduction of 1-Phenylpyridinium Chloride (12).—The procedure of Saunders and Gold[5] was followed except that 5% sodium amalgam was used and the work-up of Karrer[13] was followed. To a solution of 10.0 g (0.055 mol) of **12** dissolved in 1 l. of degassed water was added 240 g (0.53 mol) of 5% sodium amalgam. The solution was stirred for 16 hr at room temperature under nitrogen. The precipitated solid was filtered and dissolved in degassed acetone. The acetone solution was brought to the cloud point with water and cooled. Filtration of the resulting solid gave 0.60 g (7%) of **1,1′-diphenyltetrahydro-4,4′-bipyridyl (13)** as silvery plates: mp 124–126° (from acetone–water; Darco) (lit.[20] mp 136°); uv max (95% EtOH) 291 mμ (ε 38,800); nmr ($CDCl_3$) δ 7.55–6.95 (m, 10 aromatic), 6.69–6.55 (d, 4, J = 8 Hz, C-2, -2′, -6, -6′ vinyl), 4.90–4.60 (m, 4, C-3, -3′, -5, -5′ vinyl), and 3.16 (m, 2, C-4, -4′ methine).

Anal. Calcd for $C_{22}H_{20}N_2$: C, 84.58; H, 6.45; mol wt, 312. Found: C, 84.46; H, 6.60; mol wt, 309.

Additional water was added to the filtrate and the aqueous solution was extracted with ethyl ether. The ether extracts were dried (Na_2SO_4), filtered, and evaporated *in vacuo*. Recrystallization (Darco) of the residue from methanol gave 1.28 g (15%) of **6** identical with that previously reported.[5]

Registry No.—Ethyl azidoformate, 817-87-8; *p*-tosyl isocyanate, 4083-64-1; **1**, 3337-18-6; **3a**, 21471-13-6; **3b**, 21471-14-7; **4a**, 21471-15-8; **4b**, 21471-16-9; **7**, 21471-17-0; **8**, 21471-18-1; **9**, 2876-13-3; **10**, 16947-42-5; **12**, 13958-90-2.

(19) O. Magidson and G. Menschikoff, *Ber.*, 59, 1209 (1926).

(20) E. Weitz, T. Konig, and L. V. Wistinghausen, *Ber.*, **57**, 153 (1924).

Azabicyclic Alcohols. VI. Stereospecific Synthesis of the 1-Azabicylo[2.2.1]heptan-3-ol Epimers[1]

DOUGLAS O. SPRY AND HERBERT S. AARON

Chemical Research Laboratory, Edgewood Arsenal, Maryland 21010

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There are three isomeric azabicyclo[2.2.1]heptane ring systems, depending upon the position of the nitrogen atom. Recently, stereochemical studies in the 2- and 7-aza series were described.[2] In the 1-aza series,

(1) Paper V: B. P. Thill and H. S. Aaron, *J. Org. Chem.*, **33**, 4376 (1968).

(2) (a) A. Shafi'ee and G. Hite, *ibid.*, **33**, 3435 (1968); (b) P. S. Portoghese, A A. Mikhail, and H. J. Kupferberg, *J. Med. Chem.*, **11**, 219 (1968).