Experimental Section

For the potentiometric measurements, a Fisher Accumet Model 825 MP pH/mV meter was utilized together with a Corning sodium ion electrode (Cat. No. 476210) and a Corning monovalent ion electrode (Cat. No. 476220) for Na^+ and K^+ binding determinations, respectively. A Brinkman 50-mL titration vessel (Model No. EA 876-50) was utilized for all determinations. Demineralized water was prepared by passing distilled water through three Barnstead D8992 combination cartridges in series.

Melting points were taken with a Fisher Johns melting point apparatus and are uncorrected. IR spectra were obtained on neat samples (unless specified otherwise) with a Nicolet MW-S infrared spectrophotometer and are recorded in reciprocal centimeters. ^1H NMR spectra were recorded with a Varian EM 360A or EM 360 spectrometer in deuteriochloroform and chemical shifts are reported in parts per million (δ) downfield from TMS. Elemental analysis was performed by Galbraith Laboratories of Knoxville, TN.

Unless specified otherwise, reagent grade reactants and solvents were obtained from chemical suppliers and used as received. (Dibenzo-16-crown-5-oxy)acetic acid (**1**),[8] *sym*-*n*-butyl(dibenzo-16-crown-5-oxy)acetic acid (**3**),[13] and *sym*-hydroxymethyldibenzo-16-crown-5[14] were prepared by reported methods.

Preparation of *sym*-Methyl(dibenzo-16-crown-5-oxy)acetic Acid (2). Under nitrogen, 1.0 g (21.3 mmol) of NaH (60% dispersion in mineral oil) was washed with dry pentane to remove the mineral oil and was suspended in 200 mL of THF. *sym*-Hydroxymethyldibenzo-16-crown-5 (5.3 mmol) in 50 mL of THF was added and the mixture was stirred for 1 h. A solution of 1.64 g (11.8 mmol) of bromoacetic acid in 25 mL of THF was added dropwise and the mixture was stirred at room temperature for 48 h and then at reflux for 1 h. To the cooled reaction mixture was added H_2O (50 mL), and the pH was adjusted to 1 with 6 N HCl. The THF was evaporated in vacuo and the residual aqueous mixture was extracted with CH_2Cl_2 (3 × 100 mL). The combined extracts were dried ($MgSO_4$) and evaporated in vacuo to give a crude product which chromatographed twice on silica gel with CH_2Cl_2–MeOH (10:1) as eluent. Recrystallization from 30–60 °C petroleum ether–EtOAc (10:1) gave an 84% yield of **2** as a white crystalline solid: mp 102–103 °C; IR (deposited from $CDCl_3$) 3620–3200 (OH), 1730 (C=O), 1250, 1121 (CO) cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.30 (s, 3 H), 3.4–4.55 (m, 12 H), 4.75 (s, 2 H), 6.9–7.1 (m, 8 H). Anal. Calcd for $C_{22}H_{26}O_8$: C, 63.15; H, 6.26. Found: C, 62.81; H, 6.34.

Preparation of Crown Carboxylic Disulfonic Acids 4–6. A solution of Ac_2O (36 mL), AcOH (5 mL), CH_2Cl_2 (24 mL), and 2.0 g of concentrated H_2SO_4 was added to 5.0 g of crown carboxylic acid **1–3** under nitrogen. The white mixture was heated to 50 °C to produce a reddish solution which was cooled and stirred overnight at room temperature. Volatile components were removed with a rotary evaporator (heating to 45 °C). The resultant dark oil was held under vacuum (0.05 Torr) for 6 h at room temperature and then heated to 40 °C to produce off-white crystals which were dissolved in MeOH (10 mL). The MeOH solution was added to Et_2O (100 mL) and placed in a refrigerator overnight. The mother liquor was decanted and the residue was evaporated in vacuo (0.05 Torr, heating to 40 °C) to produce white crystals which were further purified by twice repeating the dissolution in MeOH, precipitation with Et_2O, and drying procedure. The final products were extremely hygroscopic white solids. Due to decomposition of **4–6** after several weeks at room temperature, they were converted into their calcium salts for storage by neutralizing an aqueous solution of the crown carboxylic disulfonic acid with 1 equiv of $CaCO_3$ and evaporation of the H_2O in vacuo to yield a white powder.

(Bis[4(5)-sulfobenzo]-16-crown-5-oxy)acetic acid (4): mp 127–128 °C; 93%; IR (deposited film from $CDCl_3$) 3408 (OH), 1734 (C=O), 1124 (CO), 1033 (S=O) cm^{-1}; ^1H NMR (CD_3SOCD_3) δ 3.5–4.5 (m, 15 H), 5.60 (s, 13 H), 6.75–6.85 (m, 6 H). Anal. Calcd for $C_{21}H_{24}O_{14}S_2 \cdot 5.0H_2O$: C, 39.51; H, 5.42. Found: C, 39.16; H, 5.33. Calcium salt: mp >300 °C dec; IR (KBr) 3400 (OH), 1603 (C=O), 1107 (CO), 1039 (SO) cm^{-1}; ^1H NMR (D_2O) δ 3.5–4.3 (m, 15 H), 6.7–7.4 (m, 6 H).

***sym*-Methyl(bis[4(5)-sulfobenzo]-16-crown-5-oxy)acetic acid (5):** mp 110–111 °C; 86%; IR (deposited film from $CDCl_3$) 3421 (OH), 1730 (C=O), 1107 (CO), 1033 (SO) cm^{-1}; ^1H NMR (CD_3SOCD_3) δ 1.40 (s, 3 H), 3.2–4.4 (m, 14 H), 6.4–7.2 (m, 6 H), 8.40 (s, 7 H). Anal. Calcd for $C_{22}H_{26}O_{14}S_2 \cdot 2H_2O$: C, 42.99; H, 4.92. Found: C, 43.07; H, 4.98. Calcium salt: mp >270 °C dec; IR (KBr) 3400 (OH), 1587 (C=O), 1100 (CO), 1039 (SO) cm^{-1}.

***sym*-*n*-Butyl(bis[4(5)-sulfobenzo]-16-crown-5-oxy)acetic acid (6):** mp 173–175 °C; 82%; IR (deposited film from $CDCl_3$) 3418 (OH), 1745 (C=O), 1113 (CO), 1035 (SO) cm^{-1}; ^1H NMR (CD_3SOCD_3) δ 0.8–1.9 (m, 9 H), 3.4–4.7 (m, 14 H), 6.7–7.3 (m, 6 H). Anal. Calcd for $C_{25}H_{32}O_{14}S_2$: C, 46.72; H, 5.04. Found: C, 46.47; H, 5.02. Calcium salt: mp >330 °C dec; IR (KBr) 3400 (OH), 1595 (C=O), 1110 (CO), 1035 (SO) cm^{-1}.

Potentiometric Determination of Stability Constants. The ion-selective electrode was preconditioned by soaking overnight in a 0.1 M solution of the alkali metal chloride in buffered solution at the pH of interest. For pH 1–6, a phosphoric acid–trimethylammonium hydroxide buffer was used, while a phosphoric acid–tris(2-hydroxyethyl)ammonium buffer was utilized for pH >6.[15] The buffer concentrations were 0.001 M in both cases and the buffer pH was measured to 0.001 pH unit. At pH 5.2 and 9.0, calibration plots of mV vs. log a were linear from 0.01 to 0.0001 M alkali metal chloride. In a typical run, the titration cell was filled with 25 mL of 0.01–0.001 M alkali metal chloride in the appropriate buffer while flushing with nitrogen. The ion-selective electrode and silver/silver chloride reference electrode were inserted into the cell and the solution was stirred magnetically for 15 min. The stirring was interrupted and mV readings were taken until a change no greater than 0.2 mV was observed during 5 min. This solution was titrated by adding weighed amounts of **1–6** (0.025–0.0025 M), stirring until all the crown compound had dissolved, and taking mV readings on the unstirred solution until a change no greater than 0.2 mV was observed during 5 min. The cell was emptied and refilled with a fresh solution of the alkali metal chloride in buffer and the mV reading was taken and averaged with that for the original solution. The difference in mV readings in the presence and absence of crown was used with Frensdorf's equations[7] to calculate K_1. The possible participation of K_2 was evaluated[7] and found to be negligible in all cases. All calculations were carried out with a program written in Microsoft Basic (v 2.0) on a Macintosh 512 K microcomputer.

Acknowledgment. This research was supported by a grant from the Ames Division of Miles Laboratories.

Registry No. **1**, 78708-41-5; **2**, 109686-79-5; **3**, 87598-63-8; **4**, 109719-22-4; **5**, 109719-23-5; **6**, 100107-44-6; *sym*-hydroxymethyldibenzo-16-crown-5, 69496-29-3; bromoacetic acid, 79-08-3.

(13) Bartsch, R. A.; Liu, Y.; Kang, S. I.; Son, B.; Heo, G. S.; Hipes, P. G.; Bills, L. J. *J. Org. Chem.* **1983**, *48*, 4864.
(14) Pugia, M. J.; Knudsen, B. E.; Cason, C. V.; Bartsch, R. A. *J. Org. Chem.* **1987**, *52*, 541.
(15) Fyles, T. M., McGavin, C. A. *Anal. Chem.* **1982**, *54*, 2103.

Ruthenium-Catalyzed Reaction of Carbon Dioxide, Amine, and Acetylenic Alcohol

Yoshiyuki Sasaki*

National Research Institute for Pollution and Resources, Yatabe Ibaraki 305, Japan

Pierre H. Dixneuf

Laboratoire de Chimie de Coordination Organique, Université de Rennes, Campus de Beaulieu, 35042 Rennes, France

Received March 5, 1987

Esters of carbamic acid have directly been prepared by the reactions of carbon dioxide and amines with 2-bromoalkanophenone,[1] epoxide,[2] alkyl halide,[3] and alk-

0022-3263/87/1952-4389$01.50/0

Scheme I

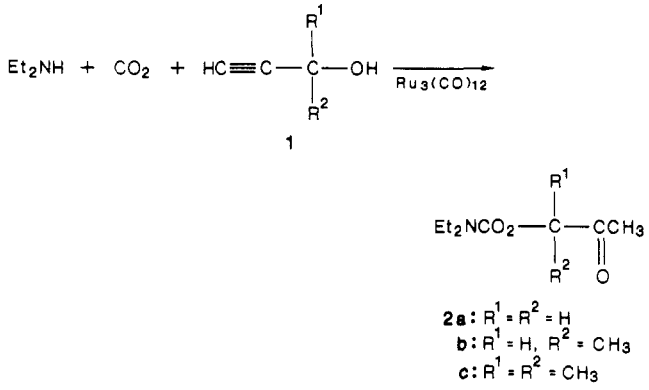


Table I. Reaction of CO_2, Amine, and Acetylenic Alcohol[a]

amine	alcohol	conv, %	product	yield, %[b]
diethylamine	**1a**	100	**2a**	54
diethylamine	**1b**	94	**2b**	64
diethylamine	**1c**	88	**2c**	52
piperidine	**1a**	85	**3**	51
morpholine	**1a**	62	**4**	4
			5	20
			6	5
n-propylamine	**1a**	67	**7a**	13
n-propylamine[c]	**1a**	93	**7a**	19
n-propylamine	**1b**	57	**7b**	28

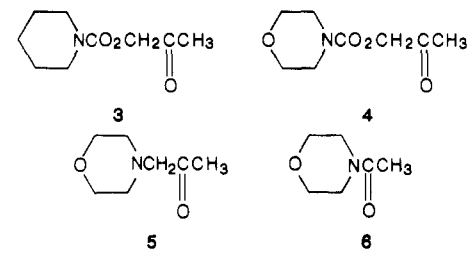
[a] Alcohol (20 mmol), amine (50 mmol), CH_3CN (5 mL), $Ru_3(CO)_{12}$ (0.2 mmol), CO_2 (50 kg/cm^2), 80 °C, 20 h. [b] Yields were determined by GLC and based on alcohols. [c] 100 °C, 20 h.

ynes.[4] We previously reported that propargyl alcohol reacted with carbon dioxide, in the presence of $Ru_3(CO)_{12}$ and Et_3N, to give carbonates involving two molecules of propargyl alcohol.[5] In this paper, it is shown that 2-oxoalkyl *N,N*-diethylcarbamates can be synthesized in one step and in moderate yields by the reactions of carbon dioxide, diethylamine, and α-ethynyl alcohols catalyzed by $Ru_3(CO)_{12}$. When *n*-propylamine is used instead of diethylamine, unsaturated oxazolidones are obtained in relatively low yields.

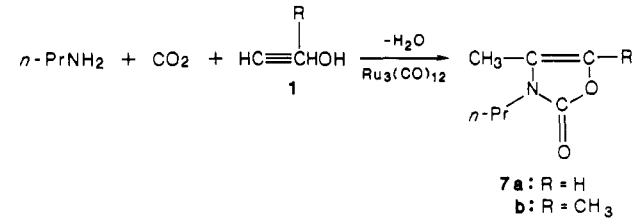
Results and Discussion

The acetylenic alcohols **1a–c** with 2.5 equiv of diethylamine in acetonitrile under a pressure of CO_2 at 80 °C afforded derivatives **2a–c** as shown in Scheme I. Regardless of the number of methyl substituents of the alcohols, 2-oxoalkyl *N,N*-diethylcarbamates **2** were obtained in more than 50% yields, while the conversion of alcohol decreased from 100% to 88% with its number of methyl groups (Table I). The similar reaction of **1a** with piperidine gave 2-oxopropyl *N,N*-pentamethylenecarbamate **3** in 51% yield. On the contrary, only 4% of carbamate **4** was formed when morpholine was used as amine, which may be due to the low basicity of morpholine. In this case the main product was amino ketone **5**, an *N*-alkylated morpholine. Acetamide **6** was also formed in a small amount (Chart I). The formation of **4** is interesting in relation with the ruthenium-catalyzed N-alkylation of amine with saturated alcohols.[6]

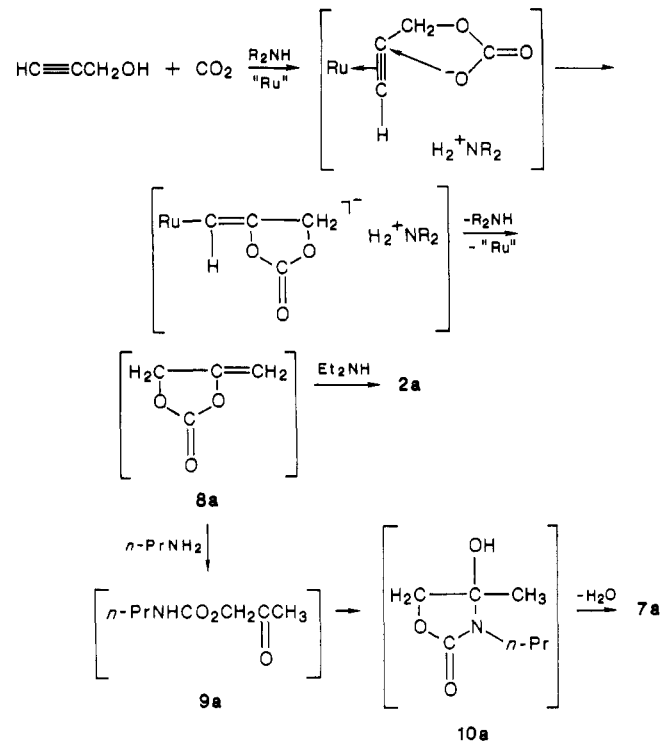
Chart I



Scheme II



Scheme III



When *n*-propylamine was used, 2-oxo-1,3-oxazolines **7** were obtained in 13–28% yields (Scheme II). The ^1H NMR spectra of **7** clearly shows an olefinic bond located in the oxazolidone ring; oxazolidones containing exocyclic double bonds are known to be formed by the reactions of 3-methyl-1-butyn-3-ol (**1c**) with CO_2 and primary amines, such as anilines, catalyzed by Cu(I) derivatives.[7]

The reaction mechanism can be rationalized as follows. α-Methylene cyclic carbonate of type **8** is obtained in the reaction of CO_2 and α-ethynyl alcohol **1b** in the

(1) Toda, T. *Chem. Lett.* **1977**, 957.
(2) Yoshida, Y.; Inoue, S. *Chem. Lett.* **1978**, 139. Yoshida, Y.; Inoue, S. *J. Chem. Soc., Perkin Trans. 1* **1979**, 3146. Asano, T.; Saito, N.; Hatakeda, K. Toda, T. *Chem. Lett.* **1978**, 311.
(3) Yoshida, Y.; Ishii, S.; Yamashita, T. *Chem. Lett.* **1984**, 1571.
(4) Sasaki, Y.; Dixneuf, P. *J. Chem. Soc., Chem. Commun.* **1986**, 790. Mahé, R.; Dixneuf, P.; Lécolier, S. *Tetrahedron Lett.* **1986**, *6333*. Sasaki, Y.; Dixneuf, P. *J. Org. Chem.* **1987**, *52*, 314.
(5) Sasaki, Y. *Tetrahedron Lett.* **1986**, 1573. Sasaki, Y. *Bull. Natl. Res. Inst. Pollut. Resour.* **1987**, *16*(4), 13.
(6) Watanabe, Y.; Tsuji, Y.; Ige, H.; Ohsugi, Y.; Ohta, T. *J. Org. Chem.* **1984**, *49*, 3259; Tsuji, Y.; Huh, K.-T.; Ohsugi, Y.; Watanabe, Y. *J. Org. Chem.* **1985**, *50*, 1365.
(7) Laas, H.; Nissen, A.; Nürrenbach, A. *Synthesis* **1981**, 958; Ger. Pat. 1 151 507.

presence of $Ru_3(CO)_{12}$ and Et_3N[5] (Scheme III). The nucleophilic addition of diethylamine to such an intermediate **8** would take place at the carbonyl to give the 2-oxoalkyl carbamate **2a**. Addition of *n*-propylamine to the intermediate **8** would normally afford the *N*-alkylcarbamate **9**. Cyclization of **9** is then followed to give the 4-hydroxyoxazolidone **10** which on dehydration would give **7**.

α-Methylene cyclic carbonate **8** from propargyl alcohol **1a** is not stable compared with those derived from **1b** or **1c**, and reacts with another molecule of **1a** to give 2-oxoalkyl carbonate[5] in the presence of Et_3N. The high yield of **2a** may be caused by the enhanced nucleophilicity of diethylamine compared with that of the alcohol **1a**, the attack of which prevents **8** from decomposition or polymerization.

Experimental Section

Acetonitrile and amines were dried over P_2O_5 and CaH_2, respectively, and distilled. The other materials were used as purchased.

In a representative reaction, a mixture of acetonitrile (5 mL), $Ru_3(CO)_{12}$ (0.2 mmol), amine (50 mmol), and α-ethynyl alcohol (20 mmol) was placed in a 100-mL autoclave and stirred at 80 °C under 50 kg/cm^2 of initial pressure of CO_2 for 20 h. The amounts of products and unreacted alcohol were determined by GLC (10% FFAP, 2 m). The typical purification method of products was as follows. The reaction solvent was distilled off under reduced pressure, and the resulting residue, dissolved in 20 mL of ether, was washed with 30 mL of dilute HCl solution (3%) several times and with water. The organic layer was dried over $MgSO_4$ for one night. The crude products obtained by evaporating ether were passed through a preparative GLC (10% FFAP, 2 m, 200 °C, 1.5 kg/cm^2 He). The isolated products were identified by using infrared, NMR, and chemical ionization type mass spectra. Elemental analysis was also performed for carbamates and oxazolidones.

2-Oxopropyl *N,N*-diethylcarbamate (2a): IR (neat) 1720 (C=O), 1690 cm^{-1} (NCOO); ^{1}H NMR ($CDCl_3$) δ 1.16 (6 H, t, CH_3, J = 7 Hz), 2.17 (3 H, s, CH_3CO), 3.41 (4 H, q, NCH_2, J = 7 Hz), 4.73 (2 H, s, OCH_2); mass spectrum, m/e 174 ($M^+ + 1$).

Anal. Calcd for $C_8H_{15}NO_3$: C, 55.47; H, 8.73; N, 8.09. Found: C, 55.45; H, 8.89; N, 8.09.

1-Methyl-2-oxopropyl *N,N*-diethylcarbamate (2b): IR (neat) 1720 (C=O), 1690 cm^{-1} (NCOO); ^{1}H NMR ($CDCl_3$) δ 1.13 (6 H, t, CH_3, J = 7 Hz), 1.34 (3 H, d, CH_3CH, J = 7 Hz), 2.12 (3 H, s, CH_3CO), 3.29 (4 H, q, CH_2, J = 7 Hz), 4.93 (1 H, q, CH, J = 7 Hz); mass spectrum, m/e 188 ($M^+ + 1$).

Anal. Calcd for $C_9H_{17}NO_3$: C, 57.73; H, 9.15; N, 7.48. Found: C, 57.50; H, 9.29; N, 7.42.

1,1-Dimethyl-2-oxopropyl *N,N*-diethylcarbamate (2c): IR (neat) 1720 (C=O), 1690 cm^{-1} (NCOO); ^{1}H NMR ($CDCl_3$) δ 1.16 (6 H, t, CH_3, J = 7 Hz), 1.46 (6 H, s, CH_3C), 2.14 (3 H, s, CH_3CO), 3.31 (4 H, q, CH_2, J = 7 Hz); mass spectrum, m/e 202 ($M^+ + 1$).

Anal. Calcd for $C_{10}H_{19}NO_3$: C, 59.68; H, 9.51; N, 6.96. Found: C, 59.38; H, 9.55; N, 7.00.

2-Oxopropyl *N,N*-pentamethylenecarbamate (3): IR (neat) 1725 (CO), 1695 cm^{-1} (NCOO); ^{1}H NMR ($CDCl_3$) δ 1.59 (6 H, m, CH_2), 2.14 (3 H, s, CH_3CO), 3.44 (4 H, m, NCH_2), 4.58 (2 H, s, OCH_2CO); mass spectrum, m/e 186 ($M^+ + 1$).

Anal. Calcd for $C_9H_{15}NO_3$: C, 58.36; H, 8.16; N, 7.56. Found: C, 58.28; H, 8.36; N, 7.86.

2-Oxopropyl *N,N*-(oxydiethyl)carbamate (4): IR (neat) 1725 (CO), 1700 cm^{-1} (NCOO); ^{1}H NMR ($CDCl_3$) δ 2.16 (3 H, s, CH_3CO), 3.56 (4 H, m, NCH_2), 3.65 (4 H, m, OCH_2), 4.67 (2 H, s, OCH_2CO); mass spectrum, m/e 188 ($M^+ + 1$).

Anal. Calcd for $C_8H_{13}NO_3$: C, 51.33; H, 7.00; N, 7.48. Found: C, 51.82; H, 7.44; N, 7.55.

4-(2-Oxopropyl)morpholine (5): IR (neat) 1715 cm^{-1} (CO); ^{1}H NMR ($CDCl_3$) δ 2.16 (3 H, s, CH_3CO), 2.45 (4 H, t, NCH_2, J = 4 Hz), 3.20 (2 H, s, NCH_2CO), 3.72 (4 H, t, OCH_2, J = 4 Hz); mass spectrum, m/e 144 ($M^+ + 1$).

4-Acetylmorpholine (6): IR (neat) 1635 cm^{-1} ($NCOCH_3$); ^{1}H NMR ($CDCl_3$) δ 2.09 (3 H, s, CH_3CO), 3.63 (8 H, m, OCH_2CH_2N); mass spectrum, m/e 130 ($M^+ + 1$).

3-Propyl-4-methyl-2-oxo-1,3-oxazoline (7a): IR (neat) 1740 (NCOO), 1655 cm^{-1} (C=C); ^{1}H NMR ($CDCl_3$) δ 0.95 (3 H, t, CH_3, J = 7 Hz), 1.68 (2 H, m, CH_2, J = 7 Hz), 2.04 (3 H, d, CH_3, J = 1.5 Hz), 3.52 (2 H, t, NCH_2, J = 7 Hz), 6.61 (1 H, q, HC=, J = 1.5 Hz); mass spectrum, m/e 142 ($M^+ + 1$).

Anal. Calcd for $C_7H_{11}NO_2$: C, 59.56; H, 7.85; N, 9.92. Found: C, 59.98; H, 8.05; N, 9.93.

3-Propyl-4,5-dimethyl-2-oxo-1,3-oxazoline (7b): IR (neat) 1740 (NCOO), 1695 cm^{-1} (C=C); ^{1}H NMR ($CDCl_3$) δ 0.95 (3 H, t, CH_3, J = 7 Hz), 1.68 (2 H, m, CH_2, J = 7 Hz), 1.96 (3 H, s, CH_3), 2.02 (3 H, s, CH_3), 3.42 (2 H, t, NCH_2, J = 7 Hz); mass spectrum, m/e 156 ($M^+ + 1$).

Anal. Calcd for $C_8H_{13}NO_2$: C, 61.91; H, 8.44; N, 9.03. Found: C, 61.57; H, 8.57; N, 9.00.

Acknowledgment. We are grateful to Dr. Y. Urushigawa of N.R.I.P.R. for GC-MS analysis.

Registry No. **1a**, 107-19-7; **1b**, 2028-63-9; **1c**, 115-19-5; **2a**, 109687-47-0; **2b**, 109687-48-1; **2c**, 91017-18-4; **3**, 109687-49-2; **4**, 109687-50-5; **5**, 6704-35-4; **6**, 1696-20-4; **7a**, 109687-51-6; **7b**, 109687-52-7; CO_2, 124-38-9; Et_2NH, 109-89-7; $PrNH_2$, 107-10-8; $Ru_3(CO)_{12}$, 15243-33-1; piperidine, 110-89-4; morpholine, 110-91-8.

Dicyclopenta[*ef,kl*]heptalene (Azupyrene) Chemistry. Electrophilic Monosubstitution: Acetylation, Halogenation, and Thiocyanation. 1-(Ethoxymethyl)azupyrene and Dimethyl (1-Azupyrenylmethyl)malonate. Acetylazupyrene Geometry[1]

Arthur G. Anderson, Jr.,* and Edward D. Daugs[2]

Department of Chemistry, University of Washington, Seattle, Washington 98195

Received May 12, 1986

In the previously reported studies on the electrophilic monosubstitution of the title compound **1** it was found that the ratios of 1- to 4-substitution were 6:1 for protonation, 13:1 for trifluoroacetylation, and ca. 1:35 for nitration.[3] An interpretation of these results based on MNDO calculations was presented. The present paper gives the experimental findings for the three additional reactions given in the title and the preparation of two other monosubstitution compounds.

Acetylation. The reaction of **1** with acetic anhydride and boron trifluoride etherate or acetyl chloride and aluminum chloride gave an 8:1 mixture of the 1-acetyl (**2**) and 4-acetyl (**3**) derivatives based on the NMR analysis of the unseparated reaction mixture. It was possible to separate these products by flash chromatography and characterize them by their ^{1}H NMR spectra. The majority of the maxima in the UV-vis spectrum of **2** corresponded to those observed earlier for the 1-trifluoroacetyl compound.[3b]

Halogenation. Treatment of a solution of **1** with bromine or with *N*-bromosuccinimide rapidly yielded a 11.5:1 mixture of 1-bromo- (**4**) and 4-bromoazupyrene (**5**) as determined by the NMR and mass spectra of the unseparated product mixture. Purification afforded a mixture

(1) Presented in part of the 39th Northwest Regional Meeting of the American Chemical Society, Moscow, Idaho, June 13-15, 1984.
(2) From: Daugs, E. D. Ph.D. Thesis, University of Washington, 1985.
(3) (a) Anderson, A. G., Jr.; Davidson, E. R.; Daugs, E. D.; Kao, L. G.; Lindquist, R. L.; Quenemoen, K. A. *J. Am. Chem. Soc.* **1985**, *107*, 1896. (b) Anderson, A. G., Jr.; Masada, G. M.; Kao, G. L. *J. Org. Chem.* **1980**, *45*, 1312.

0022-3263/87/1952-4391$01.50/0