with eventual disappearance of the nmr signals characteristic of **2b**.

Photoisomerization of 1c.—A solution of **1c**[2c] (0.50 g) in ethanol (60 ml) was irradiated using a Type L lamp for 5 hr. Evaporation of the solvent left pure **2c** having infrared and nmr spectral properties identical with those of previously characterized **2c**.[2c]

Photoisomerization of 1d.—Irradiation of **1d** (160 mg) in methanol (60 ml) for 3 hr using the Type S lamp resulted in complete disappearance of **1d** as shown by tlc. The material obtained by evaporation of the solvent had nmr and ir spectra which were very similar to those of an authentic sample of **2d**.[2c] The aromatic:vinyl integration ratio was about 7:1, however.

Preparation of 1e.—A mixture of **1c** and **2c** (80 g) prepared by the method of Sundberg, Ligon, and Lin[2a] was refluxed for 24 hr with 300 ml of 10% sodium hydroxide solution. The cooled alkaline solution was extracted with ether to remove organic impurities and then made strongly acidic with concentrated hydrochloric acid and extracted with ether to remove benzoic acid. The aqueous layer was made alkaline with concentrated sodium hydroxide and treated at 0° with small portions of benzyl chloroformate with vigorous shaking (total 40 ml). The solution was kept alkaline by addition of small portions of concentrated sodium hydroxide during the acylation. The reaction mixture was extracted with ether and acidified, and then the mixture of exocyclic and endocyclic acids was extracted with chloroform (93% yield). Crystallization from absolute ether gave the exocyclic isomer, 1-carbobenzyloxy-$\Delta^{4,\alpha}$-piperidineacetic acid: mp 127.5–128.5°; nmr ($CDCl_3$) δ 11.35 (s, 1), 7.4 (s, 5), 5.8 (s, 1), 5.2 (s, 2), 3.6 (broad t, 2).

Anal. Calcd for $C_{15}H_{17}NO_4$: C, 65.44; H, 6.22; N, 5.09. Found: C, 65.28; H, 6.30; N, 5.01.

Esterification with diazomethane gave **1e**.

Photoisomerization of 1e.—Irradiation of a solution of **1e** (8.3 g) in methanol (150 ml) for 22 hr using the Type L lamp gave after evaporation of the solvent 7.8 g of **2e** having spectral properties identical with those of an analytical sample prepared by bulb-to-bulb distillation: ir 1750 (ester CO), 1720 cm^{-1} (carbamate CO); nmr ($CDCl_3$) δ 7.35 (s, 5), 5.48 (broad, 1), 5.12 (s, 2), 3.6 (overlapping multiplet and singlet, 5), 2.98 (s, 2) and 2.1 (broad, 2).

Anal. Calcd for $C_{16}H_{19}NO_4$: C, 66.42; H, 6.62; N, 4.84. Found: C, 66.58; H, 6.78; N, 4.84.

For routine preparative work the mixture of exocyclic and endocyclic acids described in the previous experiment was esterified with diazomethane and the resulting mixture of esters was converted to pure **2e** by irradiation.

Synthesis of 1f.—A mixture of **1d** and **2d** was prepared from 1-benzoyl-3-ethyl-4-piperidone as described by Sundberg, Ligon and Lin.[2a] Conversion to **1f** was carried out as described for **1e**. The analytical sample was purified by chromatography on Florisil: ir (neat) 1720 cm^{-1} (CO, broad, overlapping carbamate and conjugated ester); nmr ($CDCl_3$) δ 7.3 (s, 5), 5.68 (s, 1), 5.11 (s, 2), 3.62 (s, 3), 1.4 (q, 2), and 0.82 (t, 3).

Anal. Calcd for $C_{18}H_{23}NO_4$: C, 68.12; H, 7.31; N, 4.41. Found: C, 68.19; H, 7.45; N, 4.47.

Isomerization of 1f.—A solution of **1f** (15.5 g, 0.049 mol) in methanol (500 ml) was irradiated with the Type L lamp for 20 hr. Evaporation of the solvent gave a residue which was eluted from Florisil F-100 (200 g) with 10% ether in benzene. Evaporation of the solvent gave a quantitative yield of material having spectral properties identical with those of the analytical sample, prepared from a center fraction: ir (neat) 1749 (CO) and 1720 cm^{-1} (carbamate CO); nmr ($CDCl_3$) δ 7.4 (s, 5), 5.5 (broad s, ~1), 5.18 (s, 2), 3.68 (s, 3), 3.08 (s, 2), 2.1 (broad q, 2) and 1.0 (m, 2).

Anal. Calcd for $C_{18}H_{23}NO_4$: C, 68.12; H, 7.31; N, 4.41. Found: C, 68.35; H, 7.35; N, 4.34.

Although there were no other indications of the presence of a second component, the vinyl proton integration was about 70% of the expected value, indicating that the product might contain up to 30% of the alternative endocyclic isomer.

Preparation of 1g.—Sodium hydride (2.1 g of 50% mineral oil dispersion) was rinsed with hexane and then covered with anhydrous ether (80 ml). A solution of triethyl 2-phosphonobutyrate[7] (12.6 g) in ether (20 ml) was added slowly. When hydrogen evolution had ceased, a solution prepared by dissolving 1-benzoyl-4-piperidone (10.15 g) in ether (100 ml) and benzene (20 ml) was added. The reaction mixture was then refluxed for 17 hr under nitrogen. The reaction mixture was filtered and the organic filtrate was dried and evaporated. Chromatography gave **1g** (10.2 g, 68%). The analytical sample was prepared by bulb-to-bulb distillation: bp 174–175° (0.1 mm); ir (neat) 1725 (ester CO), 1640 cm^{-1} (amide CO); nmr ($CDCl_3$) δ 7.5 (s, 5), 4.25 (q, 2), 3.7 (broad, 4), 2.46 (m, 6), and 1.25 and 1.0 (overlapping t, 6).

Anal. Calcd for $C_{18}H_{23}NO_3$: C, 71.73; H, 7.69; N, 4.65. Found: C, 71.94; H, 7.65; N, 4.47.

Photoisomerization of 1g.—A solution of **1g** (100 mg) in ethanol (60 ml) was irradiated for 3.5 hr using the Type S lamp. Evaporation of the solvent left **2g** having spectral properties identical with those of the analytical sample prepared by bulb-to-bulb distillation: ir (neat) 1745 (ester CO), 1640 cm^{-1} (amide CO); nmr ($CDCl_3$) δ 7.48 (s, 5), 5.60 (bs, 1), 4.15 (q overlapping m, 4), 3.60 (b, 2), 2.90 (t, 1), 2.20 (b, 2), 1.75 (broadened q, 2), 1.25 (t, 3), 0.90 (t, 3).

Anal. Calcd for $C_{18}H_{23}NO_3$: C, 71.73; H, 7.69; N, 4.65. Found: C, 71.55; H, 7.86; N, 4.50.

Base-Catalyzed Isomerization of 1c.—A solution of **1c** (40.0 g) was dissolved in ethanol (200 ml) and treated with a sodium ethoxide solution prepared by dissolving sodium metal (0.75 g) in ethanol. After stirring for 3 hr at room temperature the solution was poured into acidic brine and extracted with ether. The product obtained by drying and evaporation of solvent was shown by nmr to be a 1:2.9 mixture of **1c** and **2c** (92% yield). Separation and characterization of **1c** and **2c** have been reported previously.[2c]

Registry No.—**1a**, 28399-82-8; **1b**, 40110-55-2; **1c**, 21363-69-9; **1d**, 21363-68-8; **1e**, 40112-93-4; **1f**, 40112-94-5; **1g**, 40112-95-6; **2a**, 37123-97-0; **2b**, 40112-97-8; **2c**, 21363-70-2; **2d**, 21389-71-9; **2e**, 30338-85-3; **2f**, 40113-01-7; **2g**, 37124-04-2; triethyl phosphonoacetate, 867-13-0; 1-benzyl-4-piperidone, 3612-20-2; 1-carbobenzyloxy-$\Delta^{4,\alpha}$-piperidineacetic acid, 40113-03-9; triethyl 2-phosphonobutyrate, 17145-91-4; 1-benzoyl-4-piperidone, 24686-78-0.

Acknowledgment.—Some of the compounds utilized in this work were originally prepared in our laboratory by Dr. F. O. Holcombe.

(7) B. Ackerman, R. M. Chlodek, and D. Swern, *J. Amer. Chem. Soc.*, **79**, 6524 (1957).

Formamoylation of Some Azo Compounds and the Characterization of Reaction Products

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The free-radical addition of formamide to olefins has been reported.[1] The reaction proceeds with good yields in the presence of peroxide or ultraviolet radiation (2000–2500 Å).[2] Likewise, the addition of a number of free-radical species to azo compounds has been observed.[3] We wish to report here the first addition of the formamoyl radical to azo compounds.

Formamide adds to 1,1′-azobisformamide (ABFA) in the presence of decomposing benzoyl peroxide (BPO)

(1) (a) D. Elad and J. Rokach, *J. Org. Chem.*, **29**, 1855 (1964); (b) D. Elad, *Chem. Ind. (London)*, 362 (1962); (c) L. Friedman and H. Schechter, *Tetrahedron Lett.*, 238 (1961); *Chem. Abstr.*, **55**, 20934*d* (1961).

(2) The addition also proceeds in sunlight.[1a,b]

(3) (a) A. Jones, E. R. Morris, and J. C. J. Thynne, *J. Phys. Chem.*, **72**, 2677 (1968); (b) D. Mackay, U. F. Marx, and W. A. Waters, *J. Chem. Soc.*, 4793 (1964).

(mole ratio 30:1:0.2, respectively) to give a product identified as trisformamoylhydrazine (**1**) in 86% yield (Scheme I).

SCHEME I

$$(PhCO_2)_2 \longrightarrow 2R\cdot \quad (1)$$

$$R\cdot + HC(O)NH_2 \longrightarrow RH + \cdot C(O)NH_2 \quad (2)$$

$$\cdot C(O)NH_2 + H_2NCON{=}NCONH_2 \longrightarrow H_2NCO\dot{N}N(CONH_2)_2 \quad (3)$$

$$H_2NCO\dot{N}N(CONH_2)_2 + HCONH_2 \longrightarrow \underset{\mathbf{1}}{H_2NCONHN(CONH_2)_2} + \cdot C(O)NH_2 \quad (4)$$

No attempt was made to optimize conditions, but our results and those of other workers suggest that the chain sequence postulated is short and/or that the initial process (eq 2) leading to formamoyl radical formation proceeds in low yield. Thus, it was observed that at levels of BPO below 5 mol %, formamoylation of ABFA does not occur. That the reaction proceeds *via* a radical course analogous to that suggested for additions to olefins was demonstrated when 83% of the ABFA was recovered unchanged in a control reaction carried out in the absence of BPO. The remaining 17% was accounted for as **1** (~9%), cyanuric acid, and urea, resulting from slight thermal decomposition of ABFA.

The reaction was extended to *N,N,N',N'*-tetramethylazobisformamide (TMABFA) and gave the corresponding formamoylated product, **2**, in 32% yield. An attempt to add formamide to azobenzene under similar reaction conditions failed to yield any 1:1 adduct (**3**). This observation suggests that for

$\underset{\mathbf{2}}{Me_2NCONHN(CONH_2)CONMe_2}$ $\underset{\mathbf{3}}{PhNHN(CONH_2)Ph}$

addition of formamoyl radical to proceed, the azo function should possess a low electron density, a condition met by the azo compounds ABFA and TMABFA. The structural assignments (**1** and **2**) for the formamoylation products of ABFA and TMABFA, respectively, were based on the following rationale: (1) direct analogy to the process and products (amides) obtained in the formamide–olefin reaction;[1] (2) from reported studies[4] involving the direct examination of the radical produced from formamide and its assigned structure as $\cdot C(O)NH_2$, and (3) from chemical and physical properties of the 1:1 adducts obtained.

Structures **1** and **2**, assigned to the addition products from ABFA and TMABFA upon reaction with formamide, are supported by elemental analyses and molecular-weight determinations. In addition, the mass spectral fragmentation pattern of the 1:1 ABFA–formamide adduct is in agreement with the assigned structure **1**. Although a molecular ion (*m/e* 161) was not readily discernible, a peak at *m/e* 118 was reasonably intense and possibly represents a charged hydrazobisformamide (biurea) species. Below *m/e* 118, the fragmentation pattern of **1** was in general agreement with that of biurea.

The nmr spectrum of **1** in DMSO-d_6 exhibits four singlets (integral ratio 2:2:2:1) between δ 6.0 and 8.0. The lowest field absorption (δ 7.95, 1 H) was assigned to the hydrazo proton. The lone formamoyl group occurs as a sharp singlet at δ 6.11 and the geminal formamoyl groups as broad singlets at δ 6.97 and 7.59. Warming to approximately 40° caused the signals at δ 6.97 and 7.59 to coalesce (δ 7.17, 4 H). The process was completely reversible on cooling.

Formamoyl-substituted hydrazo compounds do not exhibit doubling of the amide proton signals under these conditions.[5] The occurrence of two signals for the geminal formamoyl groups is a consequence of rotational barriers involving the C–N–N bonds,[6] barriers to nitrogen inversion, or possibly both.[7] We have not made a thorough study of the temperature-dependent nmr spectrum of **1**. The relative importance of each of these parameters could not be established unambiguously in **1**.

By comparison, the nmr spectrum of **2** was relatively straightforward. The nmr absorption at δ 8.77 was assigned to the single hydrazo proton and that at 6.60 to the two amide protons. Signals at δ 2.88 and 2.80 were assigned to the protons of each *N,N*-dimethylcarbamoyl group.

The chemical and thermal properties of **1** were also briefly investigated and indicate a relatively facile loss of cyanic acid and formation of biurea. Treatment of **1** with aqueous alkali or diethylamine, or refluxing an aqueous solution of **1**, gave quantitative yields of biurea and products attributed to the intermediacy of cyanic acid. Heating **1** in DMSO solution at 115° resulted in the formation of biurea and cyanuric acid as major reaction products. Thermogravimetric analysis of **1** showed an initial weight loss of *ca.* 26% between 225 and 250°, which corresponds to that calculated for cyanic acid.

$$H_2NCONHN(CONH_2)_2 \xrightarrow[\text{see text}]{\Delta} \underset{\text{biurea}}{H_2NCONHNHCONH_2} + \underset{\text{cyanic acid}}{HNCO}$$

$$3HNCO \longrightarrow \underset{\text{cyanuric acid}}{\text{HOC}_3\text{N}_3(\text{OH})_3}$$

Experimental Section

Nmr spectra were obtained with a Jeolco Model JNM-4H-100 100-MHz spectrometer (TMS internal standard) and the ir spectra on a Perkin-Elmer 451 spectrophotometer. Thermogravimetric analyses (TGA) were determined in dry air at 6°/min, using an American Instrument Co. Thermo-Grav. Analyses were performed by Galbraith Laboratories, Inc., Knoxville 21, Tenn. Melting points were determined in open capillary tubes on a Mel-Temp apparatus and are uncorrected. Boiling points are uncorrected.

Materials.—1,1'-Azobisformamide (ABFA) (Aldrich Chemical Co.) [λ_{max}^{DMSO} (ϵ_{max}) 262 (1680), 423 (51.1)], azobenzene (Eastman Organic Chemicals), and benzoyl peroxide (Lucidol Division of Wallace and Tiernan) were used as received. Formamide

(4) (a) R. Livingston and H. Zeldes, *J. Chem. Phys.*, **47**, 4173 (1967); (b) H. Bower, J. McRae, and M. C. R. Symons, *J. Chem. Soc. A*, 2400 (1971).

(5) Unpublished results from this laboratory. Note also that the remaining formamoyl group in **1** appears as a sharp singlet.

(6) G. J. Bishop, B. J. Price, and I. O. Sutherland, *Chem. Commun.*, 672 (1967).

(7) M. J. S. Dewar and W. B. Jennings, *J. Amer. Chem. Soc.*, **91**, 3655 (1969); M. J. S. Dewar and W. B. Jennings, *Tetrahedron Lett.*, 339 (1970), and references cited therein.

(Fisher, reagent grade) was dried over magnesium sulfate and distilled *in vacuo;* the fraction of bp 66–67° (1.0 mm) was retained. *N,N,N',N'*-Tetramethylazobisformamide (TMABFA) was prepared by lead tetracetate oxidation of the corresponding hydrazobisformamide suspended in methylene chloride maintained at 20 ± 2°. The crude TMABFA recrystallized from hexane–benzene (5:1, v/v) melted at 111–113° (lit.[8] mp 112–113°).

Formamoylation of ABFA.—To a stirred slurry of ABFA (11.6 g, 100 mmol) in formamide (135 g, 3.0 mol) was added benzoyl peroxide (2.42 g, 10 mmol). The reaction mixture was then heated at 80 ± 2° under an oxygen-free nitrogen atmosphere for 4 hr. Additional peroxide (10 mmol) was then added and the reaction temperature was maintained for 24 hr.

Distillation of the clear, pale orange-amber reaction mixture *in vacuo* [pot temperature 85° (0.1 mm)] left a cream-colored solid residue. The residue was triturated consecutively with portions of alcohol and ether, leaving 13.9 g (81 mmol, 86.4%) of crude trisformamoylhydrazine (1), mp 214–222° dec. The crude cream-colored solid 1 was recrystallized twice from aqueous 75% alcohol and dried *in vacuo* (0.5 mm, 110°) to give an analytical sample melting at 220–225° dec.

Anal. Calcd for $C_3H_7N_5O_3$ (1): C, 22.36; H, 4.38; N, 43.47; mol wt, 161. Found: C, 22.21, 22.37; H, 4.39, 4.44; N, 43.56, 43.59; mol wt, 162 (determined cryoscopically in DMSO).

An ir spectrum (KBr) of 1 showed NH absorptions at 3475 (s), 3410 (shoulder, s), 3390 (s), 3350 (s), 3290 (s), and 3220 (m), a series of absorptions in the carbonyl region at 1695 (vs), 1660 (vs), 1630 (m), 1580 (m), and 1515 (m), and absorptions at 1360 (s), 1090 (w), 1063 (w), and 640 cm^{-1} (m).

The uv spectrum (H_2O) of 1 exhibits no maxima above 200 nm (ϵ_{200} 7000 l. mol^{-1}).

The nmr spectrum (DMSO-d_6) of 1 showed absorptions at δ 6.11 (s, relatively sharp, 2 H), 6.97 (broad s, 2 H), 7.59 (broad s, 2 H), and 7.95 (s, 1 H). Addition of D_2O to the DMSO-d_6 solution of 1 caused greatest diminution in the absorption at δ 7.95.

The mass spectrum of 1 (20 eV, 170°) exhibits the following: *m/e* (rel intensity) 118 (95), 101 (100), 86 m (4), 75 (95), 45 (11), 44 (95), 31 (97), and 18 (32).[9]

Formamoylation of TMABFA.—A stirred solution of TMABFA (7.8 g, 45 mmol) in formamide (135 g, 3.0 mol) containing 2.42 g (10 mmol) of benzoyl peroxide was heated to 80 ± 2° under an oxygen-free nitrogen atmosphere for 4 hr. Additional peroxide (10 mmol) was added and heating was continued for 15 hr.

The clear, pale orange reaction mixture was distilled *in vacuo* (pot <80°, 0.15 mm) and left an orange-amber semisolid residue that was slurried with warm methylene chloride (80 ml) and filtered. The filter cake, crude 1-formamoyl-1,2-bis(*N,N*-dimethylcarbamoyl)hydrazine (2) (3.15 g, 32%), mp 180–185°, was recrystallized twice from absolute alcohol to afford analytically pure 2: mp 183–184.5°; ir (KBr) 3430 (s, NH), 3220 (m), 2930 (w), 1690 (s, shoulder, C=O), 1675 (vs, C=O), 1690 (s, shoulder, C=O), 1365 (m), and 1272 cm^{-1} (w); uv (H_2O) exhibits no maxima above 200 nm (ϵ_{200} 14,600 l. mol^{-1}); nmr (DMSO d_6) δ 2.80 (s, 6 H, CH_3), 6.60 (s, 2 H), and 8.77 (s, 1 H). *Anal.* Calcd for $C_7H_{15}N_5O_3$ (2): C, 38.70; H, 6.96; N, 32.24; mol wt, 217. Found: C, 38.55; H, 7.01; N, 32.44; mol wt, 210 (determined in THF by vapor phase osmometry).

Hydrolysis of Trisformamoylhydrazine (1).—A clear solution of 1 (1.0 g, 6.2 mmol) in 75 mol of water was heated to reflux. After 4.5 hr, the reaction mixture contained some insolubles and ammonia was detected. The reaction mixture was refluxed for an additional 2 hr, cooled, and filtered. The dried filter cake (0.7 g) was identified as biurea (5.9 mmol) by melting point, mixture melting point, and ir. Evaporation of the weakly basic aqueous filtrate left a solid residue (undetermined amount) identified by ir as urea.

Basic Hydrolysis of Trisformamoylhydrazine (1).—To a slurry of 1 (1.0 g, 6.2 mmol) in 50 ml of water was added 5 ml of aqueous 40% sodium hydroxide, causing immediate solution of 1. After several minutes at room temperature, the reaction mixture become turbid and a finely divided solid precipitated. The filtered and dried solid (0.7 g) was identified as biurea (5.9 mmol). An acidulated aqueous solution of benzoyl hydrazide was added to the clear, pale yellow basic filtrate from the main reaction mixture. Upon cooling the mixture, 4-benzoylsemicarbazide precipitated; it was identified by melting point (222–225° dec), mixture melting point, and ir.

Aminolysis of Trisformamoylhydrazine (1).—To a suspension of 1 (1.0 g, 6.2 mol) in water (25 ml) at room temperature was added a solution of diethylamine (2.0 g, 41.0 mmol) in water (*ca.* 15–20 ml). After *ca.* 15 min, a solid precipitated from the turbid reaction mixture. After an additional 30 min at room temperature, the solid was filtered and the filter cake was washed consecutively with alcohol and ether. The dried filter cake (0.7 g) was identified as biurea (6.0 mmol) by melting point and ir.

The aqueous filtrate was evaporated to dryness, and the residual solid was extracted with ether. Concentration of the combined ether extracts gave diethylurea, mp 65–69°, identified by ir and nmr.

Thermolysis of Trisformamoylhydrazine (1).—A solution of 1 (6.5 mmol) in 25 ml of DMSO was heated at 115° for 21 hr. The solvent was removed by distillation *in vacuo* (pot temperature <80°, 0.1 mm). The solid residue was found to contain biurea (5.1 mmol) and cyanuric acid (2.1 mmol), as determined by nmr.

Registry No.—1, 39981-78-7; 2, 40081-62-7; ABFA, 123-77-3; formamide, 75-12-7; TMABFA, 10465-78-8; biurea, 110-21-4; benzoyl hydrazide, 613-94-5; 4-benzoylsemicarbazide, 39981-79-8; diethylamine, 109-89-7; diethylurea, 623-76-7; cyanuric acid, 108-80-5.

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(8) R. J. Crawford and R. Raap, *J. Org. Chem.*, **28**, 2419 (1963).

(9) The mass spectrum of authentic biurea (obtained from Aldrich Chemical Co., Inc.) exhibits the following fragmentation pattern (20 eV, 210°): *m/e* (rel intensity) 118 (2), 101 (89), 75 (100), 60 (21), 45 (85), 44 (90), 31 (90), 30 (54), 18 (82), and 17 (41).

The Electrocyclodimerization of *N*-Vinylcarbazole

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Passage of electric current through solutions of *N*-vinylcarbazole (VCZ), for example, with silver perchlorate in nitrobenzene[1] or with zinc bromide in acetone,[2] has been shown to result in polymerization. However, Breitenbach[3] also reported that electroinitiation with Hg^{II} cyanide, in acetonitrile, gave a cyclic dimer. Similarly, photoirradiation in the presence of organic electron acceptors results in the production of polymer or cyclic dimer depending on the acceptor and the solvent. Tada, *et al.*,[4] found that the basic character of the solvent was a critical factor in determining the production of cyclic dimer or polymer. In copolymerization studies with electron-accepting monomers,[5] it was found that as the concentration of the electron-accepting monomer decreased the proportion of cyclodimer product increased. It was con-

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(4) K. Tada, Y. Shirota, S. Kusadazashi, and H. Mikawa, *Chem. Commun.*, 1169 (1971).

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