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Synthesis and Properties of Fluorine-Containing Heterocyclic Compounds. V. Trifluoromethyl-1,8- and 1,10-Phenanthrolines (1)

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The condensation of ethyl trifluoroacetoacetate with various heterocyclic amines yielded 2-(trifluoromethyl)phenanthrolin-4-ones. The structures of these compounds were established from a study of their infrared and ultraviolet spectra. Substituted phenanthrolines were prepared by nucleophilic attack on a chlorophenanthroline. The oxidation of 5-methoxy-2-trifluoromethyl-1,10-phenanthroline yielded an *o*-quinone which was treated with typical quinone reagents. A molecular orbital calculation was carried out in an attempt to explain the ease of reduction of the 1,10-phenanthroline system and the colors of the reduced compounds.

Previous work in this laboratory has dealt with the condensation of ethyl trifluoroacetoacetate with *o*-phenylenediamine (2) and various aromatic amines (3,4). In the present investigation, we have extended this condensation to heterocyclic amines.

An acetoacetic ester can react with an aromatic amine to give either an acetoacetanilide or a β -aminocrotonate (5,6). The β -aminocrotonate can be cyclized to form a 4-quinolone (7,8). The acetoacetanilide can be cyclized to yield a 2-quinolone (9,10). Either the 2- or 4-quinolone may be prepared without isolating the intermediates.

We have prepared phenanthrolines by the condensation of ethyl trifluoroacetoacetate with heterocyclic amines, in polyphosphoric acid, at 140-150° (11). 5-Aminoisoquinoline yielded 2-(trifluoromethyl)-1,8-phenanthrolin-4-one (I) and 8-aminoquinaldine produced 9-methyl-2-(trifluoromethyl)-1,10-phenanthrolin-4-one (II). 5-Methoxy-2-(trifluoromethyl)-1,10-phenanthrolin-4-one (III) was previously prepared (4) from the same ester and 8-amino-6-methoxyquinoline. The identity of the products as phenanthrolin-4-ones was established from infrared and ultraviolet data. 2- and 4-Quinolones may be distinguished by their infrared absorption bands (12-14). 2-Quinolones absorb in the 1667-1641 cm^{-1} range while 4-quinolones absorb between 1630-1620 cm^{-1} . More recent work has shown that the range for 4-quinolones should be extended as far as 1550 cm^{-1} (15,16). All of the compounds prepared in this investigation absorbed in the 1630-1586 cm^{-1} range, indicating that these products had the phenanthrolin-4-one structure. This was supported by the ultraviolet spectra of these compounds which showed a minimum at 270 μ characteristic of the 4-quinolone isomer.

4-Chloro-5-methoxy-2-(trifluoromethyl)-1,10-phenanthroline (IV) was prepared from III by the action of phosphorus oxychloride and phosphorus pentachloride (4). Compound IV reacted easily with sodium ethoxide, *n*-butylamine and *N,N*-diethylethylenediamine to give compounds Va, Vb, and Vc.

Reductive dehalogenation of IV, not only removed the chloro group but also reduced one of the pyridine rings (4). We have definitely established the structure of this product as VI. The infrared spectrum of VI showed a band at 3416 cm^{-1} typical of a normal secondary amine. The mass spectrum indicated that the loss of the trifluoromethyl group from the parent compound was small. A more intense peak would have been expected had the ring which contained the trifluoromethyl group been reduced. The nmr spectrum of VI, in deuteriochloroform, confirmed its structure. A quintet centered at 7.94 τ (2 protons, $J = 6.0$ cps) was assigned to the methylene group at C-3. Two triplets centered at 7.10 τ and 6.54 τ (2 protons each, $J = 6.0$ cps) were attributed to the methylene groups at C-4 and C-2. A singlet at 6.12 τ (3 protons) was due to the protons of the methoxy group. A slightly rounded singlet at 5.04 τ (1 proton) corresponded to the N-H group of the reduced ring. A singlet was observed at 3.34 τ (1 proton) for the hydrogen at C-5. Two doublets centered at 2.38 τ and 1.38 τ (1 proton each, $J = 9.0$ cps) were due to the hydrogens at C-8 and C-7.

Because of the unexpected reduction of IV, we investigated the behavior of the parent phenanthrolin-4-one derivative (III) under similar conditions. Reduction also occurred and compound VII was obtained. The structure of VII was proven by the same methods used for compound VI.

chloride and phosphorus oxychloride, the hydroxyl group was replaced with chlorine (XIV). When XIV was treated with hydrogen and Raney nickel, ring reduction as well as the removal of the chloro group occurred. The structure of the reduced product was also established by physical methods.

The previous results showed two anomalies. Firstly, 5-methoxy-2-(trifluoromethyl)-1,10-phenanthroline-4-one (III), 4-chloro-5-methoxy-2-(trifluoromethyl)-1,10-phenanthroline (IV) and compounds XII and XIV both incorporating the 1,10-phenanthroline moiety, all underwent ring reduction when treated with hydrogen and Raney nickel, whereas other phenanthrolines are not reduced under similar conditions (4). Secondly, the reduced compounds were much more highly colored than their unsaturated precursors. We attempted to explain the first anomaly by calculating the resonance energies of 1,10-phenanthroline, 1,7-phenanthroline and their respective tetrahydro analogues using a Hückel molecular orbital calculation (18-20). Since any inclusion of substituents on these molecules was beyond the scope of this work, only the parent systems were studied. The program used was based upon the LCAO molecular orbital or Hückel theory (21). The molecules studied were 1,10-phenanthroline, 8-aminoquinoline, 1,7-phenanthroline and 7-aminoquinoline. 8-Aminoquinoline and 7-aminoquinoline were substituted for the tetrahydro-phenanthrolines corresponding to 1,10- and 1,7-phenanthrolines respectively. This simplification may be justified by the fact that only the amino nitrogens have electrons which will affect the resonance energies of these systems. Therefore the carbon atoms in the saturated ring need not be included in the calculations. The parameters used were obtained from the results of Streitwieser who has correlated reported literature values (22). Based upon these values, the resonance energy, $D\bar{E}_\pi$, for each molecule was calculated in terms of β . The following values were obtained: 1,10-phenanthroline, 5.07374; 8-aminoquinoline, 5.07266; 1,7-phenanthroline, 5.27692; 7-aminoquinoline, 5.07434. The difference between the unsaturated and reduced compound is 0.00108 for the 1,10-phenanthroline system and 0.20258 for the 1,7-system. Using an accepted value of β of -20 kcal/mole (23) we find that 1,7-phenanthroline is approximately 4.1 kcal/mole more stable than the 7-aminoquinoline while there is no measurable difference between 1,10-phenanthroline and 8-aminoquinoline. In spite of the qualitative nature of these calculations, they seem to indicate that the 1,10-phenanthroline system should be more susceptible to reduction.

The fact that the reduced 1,10-phenanthrolines absorbed at a longer wavelength, in the ultraviolet, than their aromatic precursors still had to be explained. In principle, the ultraviolet spectra of heteroatom π -systems should be calculable if appropriate h_x and k_{cx} parameters are known.

Although some limited work has been done along these lines no extensive correlations of heteroatom systems and ultraviolet spectra have been attempted (24).

In the development of the Hückel theory, it is a logical sequence that the transition between two molecular orbitals be chosen as the difference in the corresponding one-electron orbital energies. The prediction of the absorption frequencies then follows immediately from a consideration of the orbital-energy pattern. The Hückel theory, without overlap, predicts the highest wavelength band to correspond to the transition of an electron from the highest occupied molecular orbital, ψ_m , to the lowest unoccupied molecular orbital, ψ_{m+1} . Hence, we look for a correlation between the observed frequency and the energy difference between the two orbitals.

The calculations to obtain these values for 1,10-phenanthroline and 8-aminoquinoline were carried out using the identical program and parameters described in the previous section. The wavelengths or positions of absorption corresponding to these energy differences were then calculated using $\beta = 28,000 \text{ cm}^{-1}$. The value of β is obviously an arbitrary selection. The literature reports ranges from approximately $19,000 \text{ cm}^{-1}$ to $29,000 \text{ cm}^{-1}$ so our choice is in agreement with accepted values for this type of calculation (25,26). The results of these computations are summarized below.

	1,10-Phenanthroline	8-Aminoquinoline
ψ_{m+1}	-0.52712	-0.50725
ψ_m	0.61835	0.56063
ΔE	1.14547	1.06788
λ calcd.	312 $m\mu$	334 $m\mu$
λ exp't	324 $m\mu$ (24)	338 $m\mu$ (27)
λ for (III)	345 $m\mu$	(VII) 356 $m\mu$ (VI) 424 $m\mu$

While these correlations are not perfect, they are of the proper direction and magnitude. In spite of the limitations of the simplified Hückel theory, the approximations made offer possible explanations for the anomalies noted.

EXPERIMENTAL

Infrared spectra were determined on a Perkin Elmer 521 spectrometer. Ultraviolet spectra were run on a Cary 14 recording spectrometer. Nuclear magnetic resonance spectra were obtained with a Varian HR-60 spectrometer. Chemical shifts are reported as τ (p.p.m.) relative to tetramethyl silane. Microanalyses were carried out by Dr. A. Bernhardt, Max Planck Institut, 433 Mülheim (Ruhr), West Germany.

2-(Trifluoromethyl)-1,8-phenanthroline-4-one (I).

A mixture of 5-aminoisoquinoline (7.0 g., 0.05 mole) and 50 ml. of polyphosphoric acid was stirred and heated to $100-110^\circ$. Ethyl trifluoroacetoacetate (10.8 g., 0.06 mole) was added in small

portions and the mixture heated at 135-145° for 2 hours. The brown mixture was cooled in an ice bath and diluted with water (150 ml.) whereupon the yellow compound formed was removed by filtration to give 5.8 g. (45% yield) of 2-(trifluoromethyl)-1,8-phenanthroline-4-one (I). It was best purified by recrystallization once from pyridine and then repeatedly from methanol to give small white needles, m.p. 378° dec.; I showed a strong band in the infrared at 1586 cm^{-1} (C=O); ultraviolet, λ max (methanol) 332 μ (log ϵ 3.38), 306 μ (log ϵ 4.05), 295 μ (log ϵ 4.07), 245 μ (log ϵ 4.59) and 207 μ (log ϵ 4.48).

Anal. Calcd. for $\text{C}_{13}\text{H}_7\text{F}_3\text{N}_2\text{O}$: C, 59.09; H, 2.67; F, 21.57; N, 10.61. Found: C, 58.93; H, 2.80; F, 21.38; N, 10.83.

9-Methyl-2-(trifluoromethyl)-1,10-phenanthroline-4-one (II).

A mixture of 8-aminoquinoline (16.5 g., 0.10 mole) and 60 ml. of polyphosphoric acid was stirred and heated to 100-110°. Ethyl trifluoroacetate (20.2 g., 0.11 mole) was added to it in small portions. The mixture was treated as previously described for I but the resulting solution had to be neutralized with 10% sodium hydroxide to a final pH of 5 before a solid precipitated. The yellow compound was removed by filtration to give 14.7 g. (53% yield) of the phenanthroline (II). It was recrystallized from ethanol to give yellow needles, m.p. 162-165°; infrared 1618 (s) cm^{-1} , (C=O); ultraviolet, λ max (methanol) 328 μ (log ϵ 3.86), 316 μ (log ϵ 3.87), 307 μ (log ϵ 3.84), 287 μ (log ϵ 4.12) sh, 264 μ (log ϵ 4.38).

Anal. Calcd. for $\text{C}_{14}\text{H}_9\text{F}_3\text{N}_2\text{O}_2$: C, 60.43; H, 3.26; F, 20.49; N, 10.07. Found: C, 60.34; H, 3.34; F, 20.13; N, 9.93.

5-Methoxy-2-(trifluoromethyl)-1,10-phenanthroline-4-one (III).

This compound was prepared according to the procedure of Dey and Joullicé (4). It was most conveniently purified by dissolution in hot glacial acetic acid and reprecipitation with 10% sodium hydroxide and then recrystallization from methanol.

4-Chloro-5-methoxy-2-(trifluoromethyl)-1,10-phenanthroline (IV).

The compound was prepared according to the procedure of Dey and Joullicé (4) by the reaction of compound (III) with phosphorus oxychloride.

4-Ethoxy-5-methoxy-2-(trifluoromethyl)-1,10-phenanthroline (Va).

Sodium (0.20 g., 0.0087 g.-atom) followed by 4-chloro-5-methoxy-2-(trifluoromethyl)-1,10-phenanthroline (0.70 g., 0.0023 mole) was added to absolute ethanol (75 ml.) and the mixture refluxed for 3 hours. Evaporation of the ethanol and recrystallization of the residue from carbon tetrachloride followed by sublimation *in vacuo* produced 0.62 g. (86% yield) of compound Va as a white powder, m.p. 191-195°.

Anal. Calcd. for $\text{C}_{16}\text{H}_{13}\text{F}_3\text{N}_2\text{O}_2$: C, 59.63; H, 4.06; N, 8.69. Found: C, 59.54; H, 4.20; N, 8.86.

4-n-Butylamino-5-methoxy-2-(trifluoromethyl)-1,10-phenanthroline (Vb).

n-Butylamine (0.81 g., 0.011 mole) was added to 4-chloro-5-methoxy-2-(trifluoromethyl)-1,10-phenanthroline (0.86 g., 0.0029 mole) in dimethoxyethane (75 ml.) and the solution refluxed for 36 hours. The solvent was evaporated and the residue dissolved in hot, dilute acetic acid and reprecipitated with 10% sodium hydroxide to give 5.2 g. (54% yield) of compound Vb. Recrystallization from acetone gave tan needles, m.p. 210-211°.

Anal. Calcd. for $\text{C}_{18}\text{H}_{18}\text{F}_3\text{N}_3\text{O}$: C, 61.88; H, 5.19; F, 16.32; N, 12.03. Found: C, 62.04; H, 5.38; F, 16.55; N, 11.86.

4- β -Diethylaminoethylamino-5-methoxy-2-(trifluoromethyl)-1,10-phenanthroline (Vc).

N,N-Diethylethylenediamine (1.6 g., 0.014 mole) was added to

4-chloro-5-methoxy-2-(trifluoromethyl)-1,10-phenanthroline (1.0 g., 0.0034 mole) in dimethoxyethane (75 ml.) and the solution refluxed for 48 hours. The solvent was evaporated and the residue recrystallized from ethyl acetate to give 0.94 g. (74%) of compound (Vc) as yellow crystals, m.p. 154-157°. An analytical sample was prepared by synthesizing the picrate; yellow platelets, m.p. 227-229°.

Anal. Calcd. for $\text{C}_{20}\text{H}_{23}\text{F}_3\text{N}_4\text{O}\cdot\text{C}_6\text{H}_3\text{N}_3\text{O}_7$: C, 50.24; H, 4.22; N, 15.78. Found: C, 50.49; H, 4.72; N, 15.82.

Hydrogenolysis of the 1,10-Phenanthrolines and Dipyrido[3,2-*a*:2',3'-*c*]phenazines.

The reductions were carried out using Raney nickel W-2 (28) according to the procedure of Kaemmerer, Homer and Beck (29).

6-Methoxy-9-(trifluoromethyl)-1,2,3,4-tetrahydro-1,10-phenanthroline (VI).

The compound was previously prepared by Dey and Joullicé (4); ultraviolet, λ max (ethanol) 265 μ (log ϵ 4.32); visible λ max 424 μ .

6-Methoxy-9-(trifluoromethyl)-1,2,3,4-tetrahydro-1,10-phenanthroline-7-one (VII).

The product (88% yield) was recrystallized from methanol to give bright yellow needles, m.p. 162-163°; infrared, 3422 (m) cm^{-1} (N-H); ultraviolet, λ max (ethanol) 244 μ (log ϵ 4.22), 218 μ (log ϵ 4.58); visible, λ max 356 μ (log ϵ 3.78); nmr, 7.99 τ (quintet, C-3, J = 6 cps), 7.16 τ and 6.56 τ (two triplets, C-4, C-2, J = 6 cps), 6.12 τ (singlet, OCH₃), 4.90 τ (singlet, N-H), 3.46 τ and 3.02 τ (two singlets, C-5, C-8), 0.04 τ (singlet, amide proton).

Anal. Calcd. for $\text{C}_{14}\text{H}_{13}\text{F}_3\text{N}_2\text{O}_2$: C, 56.37; H, 4.39; F, 19.11; N, 9.39. Found: C, 56.46; H, 4.57; F, 19.37; N, 9.39.

6-(Trifluoromethyl)-1,2,3,4-tetrahydrodipyrido[3,2-*a*:2',3'-*c*]phenazine-8-one (XIII).

The product was recrystallized from methanol to give the product in 87% yield as a deep red powder, m.p. 198-199°; infrared, 3416 (s) cm^{-1} (N-H); ultraviolet, λ max (chloroform) 337 μ (log ϵ 3.21), 283 μ (log ϵ 3.26), 249 μ (log ϵ 3.54).

Anal. Calcd. for $\text{C}_{19}\text{H}_{13}\text{F}_3\text{N}_4\text{O}$: C, 61.62; H, 3.54; N, 15.13; F, 15.39. Found: C, 61.46; H, 3.71; N, 15.30; F, 15.55.

6-(Trifluoromethyl)-1,2,3,4-tetrahydrodipyrido[3,2-*a*:2',3'-*c*]phenazine (XV).

The product was recrystallized from methanol to give the product in 81% yield as a deep red powder, m.p. 203-204°; infrared, 3413 (m) cm^{-1} (N-H); ultraviolet, λ max (chloroform) 339 μ (log ϵ 3.15), 295 μ (log ϵ 3.22), 268 μ (log ϵ 3.52), nmr in trifluoroacetic acid, 7.56 τ (multiplet, C-2, J = 3.0 cps), 6.78 τ and 6.04 τ (two triplets, C-1 and C-3, J = 3 cps), 2.08 τ -1.42 τ (multiplet, C-7, C-10, C-11, C-12, C-13), 0.16 τ (doublet, C-8).

Anal. Calcd. for $\text{C}_{19}\text{H}_{13}\text{F}_3\text{N}_4$: C, 64.40; H, 3.70; N, 15.82. Found: C, 64.55; H, 3.88; N, 15.77.

1-Benzyl-6-methoxy-9-(trifluoromethyl)-1,2,3,4-tetrahydro-1,10-phenanthroline (VIII).

6-Methoxy-9-(trifluoromethyl)-1,2,3,4-tetrahydro-1,10-phenanthroline (1.85 g., 0.0066 mole) was dissolved in toluene (19 ml.), distilled and dried over sodium wire and pulverized sodamide (0.27 g., 0.0069 mole) was added and the mixture was refluxed for one hour and then freshly distilled benzyl bromide (1.17 g., 0.069 mole) was added dropwise. The solution was refluxed for 18 hours with the addition of more sodamide and benzyl bromide every 6 hours. The total amount of each reagent added was sodamide (0.53 g., 0.014 mole) and benzyl bromide (2.31 g., 0.014 mole). The solvent was evaporated and the residue recrystallized from

ethanol to give 1.63 g. (67% yield) of compound VIII as yellow needles, m.p. 125-126°.

Anal. Calcd. for $C_{21}H_{19}F_3N_2O$: C, 67.73; H, 5.14; F, 15.31; N, 7.52. Found: C, 67.73; H, 5.22; F, 15.46; N, 7.52.

7-Ethoxy-1-ethyl-6-methoxy-9-(trifluoromethyl)-1,2,3,4-tetrahydro-1,10-phenanthroline · Ethyl Iodide (IX).

6-Methoxy-9-(trifluoromethyl)-1,2,3,4-tetrahydro-1,10-phenanthroline-7-one (4.0 g., 0.014 mole) was dissolved in 10% sodium hydroxide (11.5 g., 0.029 mole) and ethyl iodide (8.7 g., 0.056 mole) added. The solution refluxed with stirring for two hours. Upon cooling the reaction mixture a brown layer separated and solidified. This residue was recrystallized from acetone to give 5.3 g. (74% yield) of compound IX as white needles, m.p. 182-183° dec.

Anal. Calcd. for $C_{18}H_{21}F_3N_2O_2 \cdot C_2H_5I$: C, 47.07; H, 5.14; F, 11.17; N, 5.49. Found: C, 46.87; H, 5.13; F, 11.38; N, 5.65.

Upon heating compound IX above its melting point, the ethyl iodide distilled off quantitatively to give the parent compound in 100% yield. It was purified by sublimation *in vacuo* (0.1 mm./60°) to give a yellow powder, m.p. 91-93°.

Anal. Calcd. for $C_{18}H_{21}F_3N_2O_2$: C, 61.00; H, 5.97; N, 7.91. Found: C, 60.82; H, 6.02; N, 7.82.

Reaction of 5-Methoxy-2-(trifluoromethyl)-1,10-phenanthroline-4-one (III) with Fuming Nitric Acid.

5-Methoxy-2-(trifluoromethyl)-1,10-phenanthroline-4-one (0.92 g., 0.0031 mole) was dissolved in concentrated sulfuric acid (2.5 ml.), fuming nitric acid (1.5 ml., density 1.50 g./ml.) was added and the solution was heated at 120-125° for 2 hours with stirring. The mixture was cooled and poured into water (25 ml.). 5-Methoxy-6-nitro-2-(trifluoromethyl)-1,10-phenanthroline-4-one (X) precipitated (0.35 g., 33% yield). Recrystallization from pyridine afforded light orange needles, m.p. 345-350° dec.

Anal. Calcd. for $C_{14}H_8F_3N_3O_4$: C, 49.57; H, 2.38; N, 12.39. Found: C, 49.03; H, 2.11; N, 12.28.

Neutralization of the filtrate from the preparation of compound (X) to pH 6 with 10% sodium hydroxide produced 0.45 g. (49%) of 4-hydroxy-2-(trifluoromethyl)-1,10-phenanthroline-5,6-dione (XI). Recrystallization from ethanol afforded yellow needles, m.p. 285-287° dec.; infrared, 1653 cm^{-1} (C=O), nmr in trifluoroacetic acid, 2.26 τ (singlet, C-3), 1.52 τ (triplet, J = 4.0 cps, C-8), 0.68 τ (multiplet, J = 4.0 cps, C-7 and C-9).

Anal. Calcd. for $C_{13}H_5F_3N_2O_3$: C, 53.07; H, 1.71; F, 19.38; N, 9.52. Found: C, 53.04; H, 1.61; F, 19.61; N, 9.52.

3-(Trifluoromethyl)dipyrido[3,2-a:2',3'-c]phenazine-1-one (XII).

4-Hydroxy-2-(trifluoromethyl)-1,10-phenanthroline-5,6-dione (0.47 g., 0.0016 mole) was dissolved in refluxing ethanol (65 ml.) and *o*-phenylenediamine (0.26 g., 0.0024 mole) in ethanol (10 ml.) added to it. Immediately a light yellow compound (XII) precipitated, 0.58 g. (98% yield). Recrystallization from methanol gave fluffy, yellow needles, m.p. 294-295°.

Anal. Calcd. for $C_{19}H_9F_3N_4O$: C, 62.30; H, 2.48; F, 15.56; N, 15.30. Found: C, 62.22; H, 2.43; F, 15.84; N, 15.09.

1-Chloro-3-(trifluoromethyl)dipyrido[3,2-a:2',3'-c]phenazine (XIV).

A mixture of 3-(trifluoromethyl)dipyrido[3,2-a:2',3'-c]phenazine-1-one (1.0 g., 0.0027 mole), phosphorus oxychloride (9.3 g., 0.061 mole) and phosphorus pentachloride (1.2 g., 0.0058 mole) was heated at 130° for three hours. The mixture was cooled, poured into water and the solid material collected by filtration and recrystallized from methanol to yield 0.42 g. (40% yield) of compound (XIV) as fluffy, yellow needles, m.p. 258-261°.

Anal. Calcd. for $C_{19}H_8ClF_3N_4$: C, 59.31; H, 2.09; Cl, 9.22; F, 14.81; N, 14.57. Found: C, 59.69; H, 1.96; Cl, 9.16; F, 15.09; N, 14.48.

12(13)-Chloro-3-(trifluoromethyl)quinoxalino[2',3':5,6]pyrizzino-[2,3-f][1,10]phenanthroline-1-one (XVI).

4-Hydroxy-2-(trifluoromethyl)-1,10-phenanthroline-5,6-dione (1.0 g., 0.0034 mole) was dissolved in refluxing ethanol (120 ml.) and 6-chloro-2,3-diaminoquinoxaline (0.70 g., 0.0036 mole) in 15 ml. of ethanol was added. After refluxing for 3 hours and cooling a solid precipitated. Recrystallization from 2-ethoxyethanol afforded 0.80 g. (52% yield) of compound (XIV) as an orange-brown powder, m.p. 375° dec.

Anal. Calcd. for $C_{21}H_8ClF_3N_6O$: C, 55.70; H, 1.78; N, 18.56. Found: C, 55.67; H, 2.15; N, 18.91.

Acknowledgment.

We are indebted to Dr. Frank Jordan for his generous help with the molecular orbital calculations. We thank Mr. William B. Edwards III for the nmr measurements. A research grant NsG T-69 for the years 1966-1968 from the National Aeronautics and Space Administration is gratefully acknowledged.

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ton, Indiana. It was generously supplied to us by Dr. Edward R. Thornton of this department, who made certain modifications in its design making it accessible to us.

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Received September 9, 1967

Philadelphia, Pa. 19104