yield was 6.48 g. (0.052 mole, 55%) of p-methylphenyl-silane, b.p. 22° (54 mm.).

Anal. Calcd. for $C_7H_7SiH_3$: Si, 23.0. Found: Si, 22.6.

Matrix Infrared Studies of OF Compounds. I. The OF Radical[1]

Alfred Arkell, Russell R. Reinhard, and Lewis P. Larson
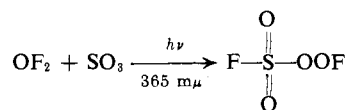
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The photolysis of OF_2 in a N_2 or Ar matrix at 4°K. has produced the OF radical. The radical has fundamental infrared absorption frequencies of 1028.5 $cm.^{-1}$ in Ar and 1025.5 and 1030.5 $cm.^{-1}$ in N_2. These frequency assignments have been confirmed in parallel runs using isotopic substitution. Some evidence is presented which explains the double absorption for OF in N_2 in terms of two types of matrix sites.

Introduction

The existence of the oxyhalide diatomic radicals, OF, OCl, OBr, and OI has been a matter of interest for over 30 years. During that time direct evidence for OCl, OBr, and OI was obtained from flame emission spectra[2] and flash photolysis coupled with ultraviolet absorption spectra.[3] The early claim for the synthesis of the OF radical by Ruff and Menzel[4] was later refuted by other workers.[5] Schumacher and co-workers[6] have investigated the photolysis and thermolysis of OF_2 and concluded that the mechanism for decomposition of OF_2 under these conditions proceeds by way of a short-lived OF radical intermediate. They were unable to detect the OF radical directly but deduced its presence from the kinetic data, quantum yield, and most convincingly from the light-induced reaction

$$OF_2 + SO_3 \xrightarrow[365\ m\mu]{h\nu} F-\underset{\underset{O}{\|}}{\overset{\overset{O}{\|}}{S}}-OOF$$

which they argued[7] must proceed by way of the OF radical. Other indirect evidence for the OF radical was obtained from electron impact studies[8] of OF_2 which indicated a low dissociation energy of about 24 kcal./mole for OF. However, values for OF dissociation derived from thermolysis studies[6c] and theoretical correlations[9,10] range much higher, 45–56 kcal./mole, indicating considerable stability for this species. Green and Linnett[11] meanwhile have argued that molecular orbital theory indicates stability for the OF^+ cation but instability for the OF radical. Previous attempts to detect the OF radical spectroscopically in order to resolve these points have failed.[3b]

The first direct and conclusive evidence for the existence of the OF radical has now been obtained using matrix isolation and infrared spectroscopy.

Experimental

All gas blends were prepared by standard manometric procedure using a matrix to reactant ratio (M/R) of 40. The OF_2 (Allied Chemical Co.), N_2 (Airco, prepurified), and argon (Airco) were used without further purification. The $O^{18}F_2$ used in some runs was prepared by the electrolysis of electrolytically dried HF containing about 0.2% of D_2O^{18}. The purified material contained approximately 91% $O^{18}F_2$ and 9% $O^{16}F_2$ according to mass spectrometric and infrared analyses.

The low temperature infrared cell is an all-metal unit similar in general design to the one described by Geiger.[12] It is a double dewar in which the central helium container is shielded from heating due to room temperature radiation by a copper radiation shield maintained at liquid nitrogen temperature (77°K.). The CsBr window on which the matrix is deposited is cooled to 4°K. by being pressed against the copper window holder which forms the bottom of the liquid helium container. Indium metal gaskets are used between the CsBr and copper for good thermal contact.

(1) This research was supported by the Research and Technology Division, AFSC, Edwards, Calif., under Contracts AF 04(611-)6083 and AF 04(611)-9577.

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(5) P. Frisch and H. J. Schumacher, *ibid.*, **229**, 423 (1936); *Z. physik. Chem.* (Leipzig), **B34**, 322 (1936); **B37** 18 (1937).

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(9) G. Glockler, *ibid.*, **16**, 604 (1948).

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(11) M. Green and J. W. Linnett, *J. Chem. Soc.*, 4959 (1960).

(12) F. E. Geiger, Jr., *Rev. Sci. Instr.*, **26**, 383 (1955).

The entire inner portion of the cell can be rotated under vacuum to bring the sample window in line with any of the four cell ports. The four ports are fitted with a gas inlet tube for deposition of gas blends, a quartz window for ultraviolet irradiation of the sample, and two silver sulfide coated silver chloride windows for infrared analysis of the sample. Teflon gaskets are used to protect the cell metal from the corrosive effect of silver chloride. Sealing of the windows, the gas inlet tube, and the rotating flange is accomplished with Viton A "O" rings.[13]

Temperature measurements are made using a gold–cobalt *vs.* copper thermocouple whose junction is sealed into a well in the side of the CsBr window. The thermocouple output is monitored by a vertical scale indicating potentiometer.[14]

Sample irradiations were made using a high pressure mercury arc, General Electric BH6. An elliptical spun aluminum reflector was used to focus the light from the arc onto the sample. The filter combinations shown in Table I were used. The liquid filters (5-cm. quartz

Results

Ultraviolet Irradiation of OF_2 in a N_2 Matrix. A series of runs was made to test the effects of various ranges of ultraviolet radiation on OF_2 decomposition.

In view of the efficient gas phase decomposition of OF_2 described by Gatti, *et al.*,[6b,7] the first test was made with monochromatic radiation of 3650 Å. After 16 min. of irradiation, there was only 2% reduction in the OF_2 absorption and no new band formation.

With the BHg-1 filter removed, leaving only the Pyrex heat shield, there was a 20% reduction in OF_2 in 10 min. and formation of a very weak doublet in the 1025–1035-cm.$^{-1}$ region. With a total of 25.5 min. of irradiation, the new bands were clearly resolved at 1027 and 1031 cm.$^{-1}$. Diffusion of the sample to 40.5°K. caused the new bands to disappear and the OF_2 absorption to increase.

After establishing that a water filter was more efficient than Pyrex in the production of the new bands, a prolonged irradiation was made with infrared spectra taken at 16–18-min. intervals. As shown in Figure 1
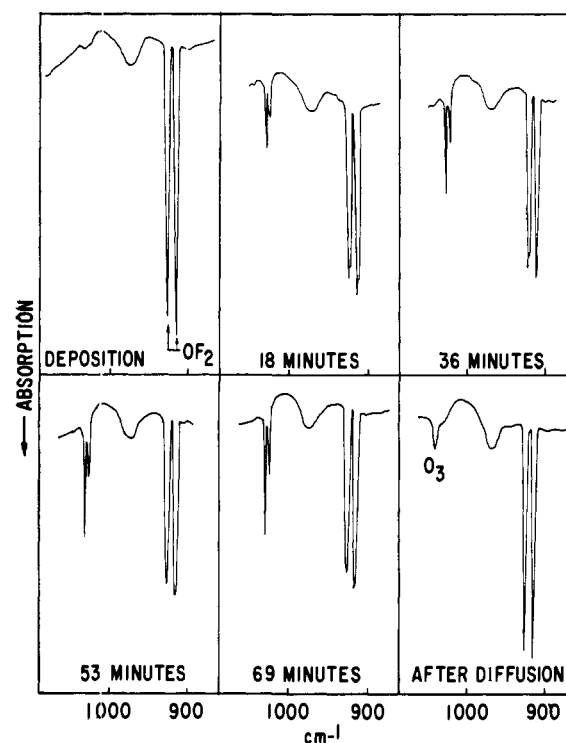
Table I

No.	Filter combination	Transmission, Å.
1	Pyrex + BHg-1	3650
2	Pyrex	3000–25,000
3	Water	2200–9000
4	Water + Corning No. 5970	3650
5	Aqueous $CuSO_4$ solution	3200–5800
6	Aqueous $NiSO_4$ solution	2200–3500 and 4500–5900

cell) were prepared according to Kasha.[15] The glass filter (BHg-1) was not adequately protected from being overheated and subsequently cracked. By using water in a quartz cell instead of a Pyrex plate as a heat shield for the Corning No. 5970, it was found that prolonged irradiations could be made.

The infrared spectra were recorded on a Beckman IR-9 spectrometer. This instrument, which is capable of 0.25-cm.$^{-1}$ resolution in the 400–4000-cm.$^{-1}$ range, had an estimated resolution and frequency accuracy of ±2 cm.$^{-1}$ for all of the runs excepting the isotopically labeled ones in which the resolution was ±1 cm.$^{-1}$.

In a typical run the blends were deposited at a rate of about 110 to 120 µmoles of mixture/min. at 4°K. Depositions usually required about 30 min. Ultraviolet irradiation periods up to 28 min. were used depending on the range of radiation being transmitted by the filters. Infrared spectra were recorded before and after deposition of the sample, and after each period of ultraviolet irradiation. At the end of each run the sample was allowed to warm to 40–45°K. (depending on whether N_2 or Ar was used as the matrix) and then cooled to 4°K. This process, termed diffusion, allows the reactive species in the matrix to migrate and react. An infrared spectrum before and after diffusion therefore gives information on the new species and the products derived from them.

(13) Obtained from C. E. Conover, Inc., Hasbrouck Heights, N. J.
(14) Obtained from Cryogenics, Inc., Alexandria, Va.
(15) M. Kasha, *J. Opt. Soc. Am.*, **38**, 929 (1948).

Figure 1. Ultraviolet irradiation of OF_2 in a nitrogen matrix at 4°K.

there was a steady increase in the new bands at 1027 and 1031 cm.$^{-1}$ during the decomposition of OF_2. The indication of an approaching equilibrated state is more readily seen when the infrared absorbances are plotted as a function of irradiation time. The resulting curves in Figure 2 clearly show the approach of a steady-state system for both OF_2 dissociation and new band formation. An additional eight infrared spectra during a 57-min. period following irradiation showed no changes in any of the absorptions. After diffusion of the sample (42°K.), the new bands disappeared and

the OF_2 absorbance increased from 0.120 to 0.168. A recovery of OF_2 of this magnitude would require a trapped radical concentration of about 0.6% basis total material in the matrix.

Using a different filter system (filter 4) to isolate the 3650-Å. radiation, a second attempt was made. The OF_2 decomposition was very slow and the production of the new bands was only moderate even after 83 min. of irradiation.[16]
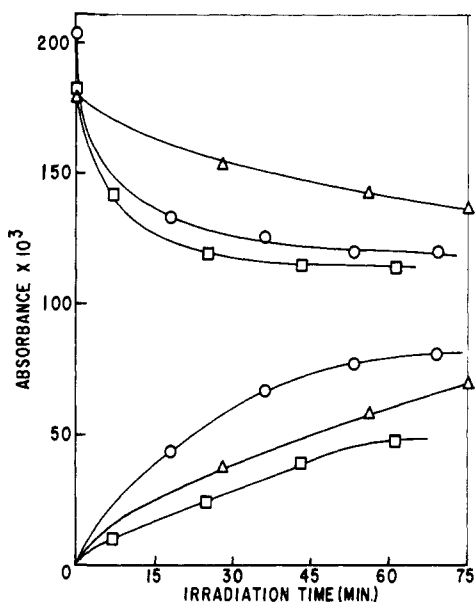

Figure 2. Ultraviolet irradiation of OF_2 in a nitrogen matrix. Filters: $\bigcirc$ water, $\square$ $CuSO_4$, $\triangle$ $NiSO_4$; upper curves, OF_2 (918 cm.$^{-1}$); lower curves, new band (1031 cm.$^{-1}$).

A rapid decomposition of OF_2 was obtained with filter 5 with equilibration after about 45 min. of irradiation (Figure 2). The new bands leveled off at about the same point in time but at a much lower level than when the water filter was used.

As shown in Figure 2, the OF_2 dissociation was much slower using filter 6, and the new band formation was intermediate between the other two filters shown. The OF_2 decomposition curve did not level off even after 2 hr. of irradiation (not shown), but the curve for the new band did. As before, the new bands were lost on diffusion, but there was no increase in the OF_2 absorbance. Instead, however, there was a small formation of O_2F_2.

Ultraviolet Irradiation of OF_2 in Argon Matrix. Irradiation of OF_2 in an argon matrix using a water filter caused OF_2 dissociation but at a faster rate than in the case of a nitrogen matrix and with no indication of equilibration. Also, only one new band was formed at 1029 cm.$^{-1}$. On diffusion the new band was lost and the OF_2 absorbance increased.

Ultraviolet Irradiation of $O^{18}F_2$ in Nitrogen and Argon Matrices. As shown in Table II, a more accurate determination of the new bands gave the values 1030.5 and 1025.5 cm.$^{-1}$ (in N_2) and 1028.5 cm.$^{-1}$ (in argon).

(16) The presence of SiF_4 as a trace impurity in this run showed, after subsequent irradiation, that the SiF_4 absorptions were unaffected by ultraviolet irradiation.

On the assumption that the new bands were due to the OF radical, a calculation for the frequency shift due to mass effect was made on that basis.[17]

Table II. Absorptions[a] Produced During the Photolysis of OF_2 at 4°K.

Matrix	$O^{16}F_2$	$O^{18}F_2$	$\Delta\nu_{obsd}$	$\Delta\nu_{calcd}$[b]
N_2	1030.5	999.5	31.0	31.6
	1025.5	994.5	31.0	31.4
Ar	1028.5	997.4	31.1	31.5

[a] In cm.$^{-1}$. [b] Calculated for the frequency shift due to mass change in going from $O^{16}F$ to $O^{18}F$. See also ref. 17.

In the nitrogen matrix the $O^{18}F$ bands were lost on diffusion, but there was no increase in the $O^{18}F_2$ absorption. In the argon matrix additional spectra, taken after the irradiation was stopped, showed that the absorbances for both $O^{18}F$ and $O^{18}F_2$ decreased. This, in addition to the lack of recovery of $O^{18}F_2$ or any $O^{18}F$-containing compound on diffusion suggested that the $O^{18}F$ radical was decomposing on warmup.

Ultraviolet Irradiation of OF_2 and N_2O in a Nitrogen Matrix. A mixture of OF_2 + N_2O + N_2 (1:2:40) was irradiated to test the effect of added N_2O on the new bands at 1025.5 and 1030.5 cm.$^{-1}$. A large increase in the 1025.5-cm.$^{-1}$ absorption compared to the 1030.5-cm.$^{-1}$ absorption made it appear that only one band at 1025.5 cm.$^{-1}$ was present. However, with improved resolution the two bands were resolved and the absorbance of the 1025.5-cm.$^{-1}$ band was found to be about 2.5 times that of the 1030.5-cm.$^{-1}$ band. In contrast to this, other runs in which no N_2O was added showed the 1030.5-cm.$^{-1}$ absorbance to be about 1.9 times that of the 1025.5-cm.$^{-1}$ band.

Discussion

The behavior of these new absorptions produced by the photolysis of OF_2 is characteristic of radicals in general: that is (1) steady formation of the new bands during the decomposition of the parent compound; (2) attainment of a limiting concentration which is of the same order of magnitude as other radical systems; (3) loss of the new bands during diffusion with recovery of some of the parent compound or with recovery of the radical dimer.

All of these were observed in this work and therefore constitute the basis for the assignment of the new bands to the OF radical. The isotopic work showed that the three new bands (1030.5 and 1025.5 cm.$^{-1}$ in N_2, 1028.5 in Ar) were shifted by the mass change indicating that all three bands were produced by oxygen-containing species. The excellent agreement between the observed band shifts and the shifts calculated for the OF radical shown in Table II gave final confirmation of the assignment of these new bands to the OF radical in nitrogen and argon matrices.

(17) The calculations for isotopic shift were made using the observed frequencies, ν_i. According to G. Herzberg, "Infrared and Raman Spectra," D. Van Nostrand Co., Inc., New York, N. Y., 1945, p. 228, this is acceptable since in general the anharmonic constants x_{ik} are small and the observed fundamentals ν_i will therefore represent fair approximations to ω_i.

The appearance of the OF radical as a doublet in N_2 and a single absorption in argon was considered unusual until it was found that other workers had made similar observations involving multiplets. In a comprehensive paper by Pimentel and Charles,[18] the observation of multiplets was discussed in terms of their three most probable causes: aggregation, rotation, and multiple sites. Although the present work did not exclude the possibility that the OF doublet in nitrogen was caused by aggregation, it was assumed that if aggregation was the cause then the OF doublet should have been observed in both the nitrogen and argon matrices. The reason for this is simply that both runs were made at the same concentration and the effect of aggregation is concentration dependent. With respect to the doublet having been caused by rotation, the statements of Pimentel and Charles[18] would seem to rule out this possibility. Of the many molecules studied, they have never found any evidence for rotational freedom in a nitrogen matrix, whereas two examples of rotation in an argon matrix are known. It seemed most unlikely therefore that the reverse situation would be found in the present work. In view of these facts, and because of a similarity with past work,[19] the possibility of a multiple site effect was given first consideration. In a search for the cause of a multiple site system in the present work, it was found that the OF doublet was most prominent in those runs in which N_2O had been generated during photolysis. It was assumed that the atomic oxygen, produced by photolysis of the OF radical, reacted with matrix nitrogen[20] to give N_2O. Nitrous oxide formed from matrix nitrogen would therefore not be expected in the argon matrix work. The results of the run in which N_2O was added to the matrix clearly showed a pronounced increase in the 1025.5-cm.$^{-1}$ absorption and a slight decrease in the 1030.5-cm.$^{-1}$ absorption. This result suggested that the 1025.5-cm.$^{-1}$ band was due to the OF radical in a nitrogen cell containing N_2O, and that the 1030.5-cm.$^{-1}$ band was due to the OF radical in a nitrogen cell containing no impurity. Further work in the general area of matrix additives is in progress and will be the subject of another paper.

The results of the photolysis with 3650-Å. radiation and the data in Figure 2 show that the effects of a matrix and the range of radiation used are the most dominant factors governing stabilization of intermediates. The notably lower efficiency of radical stabilization using 3650-Å. radiation in the present matrix work, compared to the efficient gas phase radical production described by Gatti, et al.,[6b,7] must be ascribed to a matrix phenomenon known as the cage effect. In general, the cage effect will prevent the isolation of a radical if the secondary fragment is also a radical and too large to escape the matrix cage. However, isolation will not be prevented if the secondary fragment is a neutral molecule[21] or is an

atom having sufficient residual energy to escape the cage. In the present work the fluorine atoms generated in the photolysis of OF_2 with 3650-Å. radiation apparently have low residual energy and relatively few of them escape the cage.

As the range of radiation was broadened and shifted to include a greater proportion of the region below 3650 Å., the efficiency of radical stabilization became much improved. The improvement, however, was not always without complication and some anomalous behavior was found. For example, using the $NiSO_4$ filter, observations made during both the photolysis and diffusion stages suggested that atomic fluorine from OF_2 decomposition was being lost through some alternate mechanism. In the absence of available atomic fluorine during photolysis, no OF + F recombination could occur and all of the OF_2 would ultimately be decomposed. In the absence of atomic fluorine during diffusion, only O_2F_2 could be produced from the available OF radicals. Anomalous behavior was also found in the argon matrix work and in most of the isotopic substitution work, some of which indicated that decomposition of OF_2 and OF was occurring even after the irradiation had been stopped. Although insufficient work has been done to resolve this point, it would appear that the energy required for this further decomposition may reside in the molecules as excess vibrational energy. The inability of an argon matrix to dissipate this energy may account for the observed decomposition.

There are two other points of interest, the first correlating the position of the OF radical absorption with its nitrogen counterpart, and the second relating its position with respect to other known O–F stretching fundamentals. Recent work by Milligan and Jacox[22] gave the absorption for the NF radical in an argon matrix as 1115 cm.$^{-1}$. A comparison of this value with the OF radical absorption in argon at 1028 cm.$^{-1}$ gave a difference of 87 cm.$^{-1}$ which correlates well with the differences found by Harmony and Myers[23] for a similar comparison between NF_2 and OF_2. The differences between the corresponding vibrational fundamentals are shown in Table III.

Table III. Frequency Comparisons (cm.$^{-1}$)

	NF_2	OF_2	$\Delta\nu$
ν_1	1070	929	141
ν_2	573	461	112
ν_3	931	826	105
	NF	OF	
	1115	1028	87

The second point of interest concerns the position of the OF radical absorption with respect to the other known OF stretching fundamentals. The OF radical absorption at 1028 cm.$^{-1}$ in argon is the highest O–F stretching frequency of all OF-containing compounds including OF_2. This indicates, at least qualitatively, that the O–F radical bond energy may be higher than the average OF bond energy in OF_2. Although this

(18) G. C. Pimentel and S. W. Charles, Pure Appl. Chem., 7, 111 (1963).
(19) Suggested by D. E. Milligan, National Bureau of Standards, Washington, D. C., based on his own observations during the photolysis of XN_3-type molecules in nitrogen and argon matrices.
(20) W. B. DeMore and N. Davidson, J. Am. Chem. Soc., 81, 5869 (1959), observed the formation of N_2O from atomic oxygen and molecular nitrogen during the photolysis of O_3 in a nitrogen matrix at 20°K.
(21) ClN_3 in argon ($h\nu$) → ClN + N_2 in argon: D. E. Milligan, J. Chem. Phys., 35, 372 (1961).
(22) D. E. Milligan and M. E. Jacox, ibid., 40, 2464 (1964).
(23) M. D. Harmony and R. J. Myers, ibid., 37, 640 (1962).

disagrees with a mass spectrometric estimate[5] of the relative bond strengths of OF and OF_2, it agrees in general with two other theoretical estimates.[9,10] It also agrees in principal with the comparison made by Porter[3a] between Cl_2O and the ClO radical in which he points out "the influence of the odd electron in strengthening the bond." In the case of the Cl–O bond energy, the increase is 16 kcal./mole over the average Cl–O bond energy in Cl_2O. In view of this and the relative position of the OF radical absorption, it appears not too

unlikely that the OF radical bond energy will be close to Glockler's first estimate[9] of about 56 kcal./mole. It would seem then that the inability to find the OF radical in past gas phase work may be related to extreme chemical reactivity rather than to instability.

Acknowledgments. We are grateful to Dr. Robert Y. Heisler, Dr. J. M. Caffrey, Jr., Dr. Kenneth H. Ludlum, Dr. Louis C. Roess, and various members of the Fundamental Research Section for helpful discussions throughout the course of this work.

Sulfur–Nitrogen and Oxygen–Nitrogen Bond Dissociation Energies of Some N-Fluorinated Amines

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Contribution from the Gorgas Laboratory, Rohm and Haas Company, Redstone Arsenal Research Division, Huntsville, Alabama. Received August 28, 1964

The mass spectra and appearance potentials for the principal ions of FSO_2NF_2, $(FSO_2)_2NF$, SF_5NF_2, and FSO_2ONF_2 were measured. The values for $D(S-N)$ were found to be 39, 48, and 32 kcal./mole, respectively, and $D(O-N)$ for the latter compound was found to be 35 kcal./mole.

Introduction

A number of compounds containing sulfur, nitrogen, and fluorine have been reported.[1] However, until recently, no single bonded S–N or O–N N-fluorinated amines were known. The preparation of fluorosulfuryldifluoramine, FSO_2NF_2,[2] bis(fluorosulfuryl)fluoramine, $(FSO_2)_2NF$,[3] pentafluorosulfur difluoramine, SF_5NF_2,[4–6] and N,N-difluorohydroxylamine O-fluorosulfate, FSO_2ONF_2,[7] made it of interest to investigate the mass spectra and appearance potentials of the principal ions in these compounds to determine the S–N and O–N bond dissociation energies.

Experimental

The compounds were prepared by the methods reported in the literature.[2–7] Samples were purified by vacuum line fractionation followed by gas partition chromatography except for the FSO_2NF_2. It was purified only by the literature method[3] and was found to contain 4–5% SO_2. The FSO_2ONF_2 contained only a trace of SiF_4 and the $(FSO_2)_2NF$ and SF_5NF_2 showed

no impurities. Gas chromatographic, infrared, and mass spectral analyses were used for detection of impurities. The impurities found are not closely related to the compounds studied and are unlikely to cause interference with the appearance potential measurements. The shape of the ion efficiency curves did not indicate interferences. The mass spectra and appearance potentials were measured using a Consolidated Systems Corporation high speed mass spectrometer. All mass spectra were obtained at an ionization voltage of 70 v. The appearance potentials were obtained by the vanishing current method with argon added as a voltage standard. The repeller voltage was adjusted to its minimum value, the accelerator voltage was maintained constant, and magnetic scanning was used to keep the effects of contact potentials and field penetration the same during measurements. The mass spectra are shown in Table I and the appearance potentials (A.P.) in Table II. The errors shown for the appearance potentials are the estimated errors in the measurements. Table III shows the bond energies and heats of formation with the estimated error in the heats of formation.

Results and Discussion

The mass spectra obtained for these compounds are given in Table I. The mass spectrum for SF_5NF_2 differs from that reported by Cady and co-workers,[4] but it is very similar to that reported by Logothetis, *et al.*[5] The reason for this difference is not readily apparent but may be a result of the mass spectrometer source heater being turned off in this research, whereas the source heater was set at 175° when the former group recorded their spectrum.[8] The heated source may cause sample decomposition.[4] Samples generally gave more reproducible spectra if the source was unheated.

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(7) M. Lustig and G. H. Cady, *ibid.*, 2, 388 (1963).
(8) G. H. Cady, private communication.