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Chemonuclear Synthesis of Nitrogen-Fluorine Compounds

R. N. HAZLETT

*Fuels Branch
Chemistry Division*

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U.S. NAVAL RESEARCH LABORATORY
Washington, D.C.

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ABSTRACT

Mixtures of nitrogen and fluorine containing uranium-235 as uranium hexafluoride have been exposed to the radiation field of a nuclear reactor. The fission fragments formed by reaction of ^{235}U with the thermal neutron component of the reactor radiation have caused the formation of nitrogen fluorides. Those identified were nitrogen trifluoride, cis-difluorodiazine, and trans-difluorodiazine. Nitrogen trifluoride is the major product, and the amount formed is dependent upon the total energy deposited. The other two products are in equilibrium with each other, and the amount formed decreases as the radiation intensity increases. The total G value for compound formation is less than 1 molecule per 100 electron volts.

PROBLEM STATUS

This is an interim report; work on this problem is continuing.

AUTHORIZATION

NRL Problem C01-05
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CHEMONUCLEAR SYNTHESIS OF NITROGEN-FLUORINE COMPOUNDS

INTRODUCTION

The advantages to be gained from use of rocket propellants of higher energy content has stimulated intensive searches for new synthesis techniques in addition to the efforts expended to produce specific chemicals. One synthesis technique is the use of ionizing radiation because of the great amount of energy associated with each particle or ray. High energy particles which have received increased study recently are the fission fragments formed from the splitting of a fissionable nucleus in a thermal neutron field.

The fragments formed by the fission of a uranium-235 nucleus are highly charged, heavy particles with energies in the 65 to 100 Mev range. These fragments are similar to other particles used in radiation chemistry, differing from the widely used nuclei such as protons and alpha particles in the properties of charge, mass, and energy. The advantages of radiation chemistry with lighter particles (rapid thermal quenching and control of the amount and site of energy deposition) are also advantages of studies with fission fragments. Fission fragments, use of which has been termed chemonuclear chemistry (1), have two characteristics which set them apart from other forms of radiation. Fission fragments, which have a short range because of their high mass and high charge, deposit a great amount of energy in a short track. Thus, the energy deposition rate on a localized scale or linear energy transfer (LET) is very high. The second characteristic results from the high thermal neutron fluxes available in nuclear reactors. A high fission rate and hence a high energy deposition rate on a gross scale can be attained.

Early studies in the chemonuclear field have been reviewed by Dawson et al. (2), and the recent work has been detailed by Brookhaven National Laboratory (3). Major efforts have been devoted to the production of two widely used liquid propellants, nitrogen tetroxide (4-6) and hydrazine (7,8). A recent experiment (9) found that nitrogen trifluoride, a potential high energy propellant, can be synthesized from a nitrogen-fluorine mixture under the influence of fission fragments.

Previous experiments in chemonuclear radiation of gases have utilized a two-phase system, fission in a solid phase such as uranium oxide and reaction in the gas phase. The fragment loses some of its energy in the solid before reaching the gas phase, and much of the energy is not available where it is desired. The amount of energy transferred to the gas from a fission event in the solid phase is difficult to measure directly. Efforts to calculate this amount or to measure it indirectly are not always successful and leave considerable uncertainty in the radiation efficiency. The experiment in which nitrogen trifluoride was synthesized (9) is an example of this. Calculations of the energy transfer from the solid fissionable source used, UF_4 , indicated 10% of the energy was deposited in the nitrogen-fluorine gas mixture. An indirect measurement (10) gave a value of 2%.

The present study attempted to avoid this difficulty by using a homogenous fission source, uranium hexafluoride. This compound has a vapor pressure of 160 mm Hg at 25 °C, which permits a sufficient amount of the compound to be in the gas phase to give reasonable fission rates.* The gaseous system studied was a mixture of nitrogen,

*Since some decomposition of UF_6 was found in the current work, the energy deposited was not entirely from a homogeneous source.

fluorine, and uranium hexafluoride. Five compounds composed of nitrogen and fluorine are known: nitrogen trifluoride, NF_3 (11); fluorine azide, FN_3 (12); cis-difluorodiazine, N_2F_2 (13); trans-difluorodiazine, N_2F_2 (13); and tetrafluorohydrazine, N_2F_4 (14). The two N_2F_2 isomers can interchange rapidly with each other at elevated temperatures (15). The equilibrium mixture contains about 13% of the trans form at 285°C . Fluorine azide decomposes readily to the N_2F_2 isomers, and cis-difluorodiazine decomposes slowly at room temperature to N_2 and F_2 . The other N-F compounds are stable at the pressures and temperatures of interest in this work. It seems likely that if any of the N-F compounds, with the exception of fluorine azide, were formed by chemonuclear synthesis, they could be detected after a cooling period of several days.

A limited number of radiation experiments were performed with a cobalt-60 source. This permitted an evaluation of the effects of two widely differing types of radiation on product yields and ratios. The gamma-ray exposures also permitted a widespread examination of the influence of radiation dose rate, and important factor in these studies.

Combined gamma-ray thermal neutron synthesis was also explored by the use of pile radiation. In these runs, UF_6 was not included in the reaction mixture. Energy deposition is difficult to determine with mixed radiation from a pile; hence these were qualitative experiments.

EXPERIMENTAL CONSIDERATIONS

Apparatus and Experimental Procedures

The radiations were made in cylindrical aluminum cells of approximately 100-cc capacity (5-cm I.D., 0.6-cm wall, 5-cm height). Aluminum is satisfactory for use with gaseous fluorine at modest temperatures and is excellent for radiation studies in a neutron field because of the low cross section for thermal neutrons and the short half-life of ^{28}Al . Alloy 5052, which is low in copper, was suitable for cell construction. The only fitting to a cell was a needle valve fabricated from aluminum or stainless steel with Teflon packing. The valve was attached so that the packing was not exposed to the interior of the cell during radiation exposure. Cells and valves were passivated with fluorine gas prior to use. The uranium hexafluoride was loaded into the cell first; it was condensed into the cell at -78°C at the desired pressure from a known volume of the gas. After warming the cell to room temperature, fluorine was introduced to the desired pressure, followed by nitrogen.

The uranium hexafluoride was 93% enriched in the 235 isotope. The UF_6 sample container was cooled to -78°C and evacuated to remove volatile contaminants. Fluorine was passed through a sodium fluoride trap and a liquid oxygen bath. Nitrogen was dried with molecular sieves and also passed through a liquid oxygen bath. The liquid oxygen bath removed the higher C-F compounds in the fluorine and reduced the CF_4 concentration to 0.03%. This procedure also lowered the sulfur hexafluoride content of the material

The radiation cells were exposed in the pool of the NRL research reactor. During operation, the pool temperature is 30° to 35°C . The cells were mounted on an aluminum stand, which permitted exposure at various distances from the reactor core. The thermal neutron flux was altered in this way. Actual experiments were carried out in the flux range of 10^{10} to 10^{11} neutrons/cm²-sec. The period of exposure to the neutron flux varied from 1 to 7 hours. The radiated cell was then set aside in the pool for 1 to 3 days to reduce the level of radioactivity.

All experiments were performed at a total pressure of 4 atmospheres absolute. The $N_2:F_2$ ratio was normally 1:1, but one experiment at a ratio of 3:1 and another at a ratio of 1:3 were made. UF_6 pressures up to 0.1 atmosphere (≈ 0.1 g of ^{235}U) were utilized. Calculations indicated that amounts of ^{235}U greater than this would result in temperature buildup in the gas phase at thermal neutron fluxes of 10^{11} neutrons/cm²-sec.

Experiments were made to evaluate the contribution of the combined gamma-ray and thermal neutron flux from the nuclear reactor pile to the product formation. Uranium hexafluoride was not included in the reaction mixture of these blank runs, but exposure conditions were comparable to those utilized in other experiments.

Analysis

Following the cooling period, a radiated cell was attached to a copper vacuum system. The UF_6 was retained in the radiation cell by cooling to $-78^\circ C$, the majority of the sample being transferred to a 300-cc storage cell. This served as a sample source for direct analysis by gas chromatography. Alternately the products could be concentrated by pumping through a $-196^\circ C$ bath. This concentrate was then analyzed by gas chromatography. Trapping the GC (gas chromatographic) peaks permitted positive identification of the products by mass spectrometry or infrared analysis.

Initial GC analyses were with an alumina column. When it was shown that separation of N-F from C-F compounds was required, a column with 15% Kel-F 8126 oil* on alumina was found to be adequate. A 20-ft, 3/16-in.-O.D. copper column separated NF_3 from CF_4 at $-20^\circ C$. Concentrations of products were determined by comparison of GC peak areas with those for calibrated samples.

In addition to N-F compounds, small amounts of CF_4 , C_2F_6 , C_3F_8 , SF_6 , and N_2O were usually found by GC analysis. The C-F compounds were present in the fluorine feed and could be reduced but not eliminated by proper precautions. The SF_6 probably resulted from the abstraction of sulfur from the copper vacuum system, while the nitrous oxide may have been formed during the radiation by reaction of oxygen present in the fluorine gas with nitrogen.

Cobalt-60 Radiations

Gamma-ray exposures were made in the aluminum cells previously described. A 1:1 $F_2:N_2$ mixture was utilized at a total pressure of 4 atmospheres. The exposure intensity, which was determined on the basis of previous ionization chamber measurements in air, varied from 1.45×10^4 to 1.60×10^6 roentgens/hr. The absorbed dose was calculated from the exposed dose on the basis of the absorption coefficients of nitrogen and fluorine (16). Length of exposure varied from 5 to 113 days.

Energy Deposition Considerations

The amount of energy deposited in the gas phase by the fission fragments was determined by indirect means. Gold foil activation was used in measuring the thermal neutron flux (see Appendix). The number of fission events can be found from this value in conjunction with the number of ^{235}U nuclei, the thermal neutron cross section for fission, and the period of exposure (17). The total energy is calculated from this by multiplying by the energy per fission (170 Mev), which is localized on the fission fragments.

*Trademark, Minnesota Mining and Manufacturing Co.

A part of the fission fragment energy available in a homogeneous chemonuclear study is lost in the walls, since some of the fragments formed in fission events near the wall impinge in the containment vessel before transferring their energy to the gas phase. This loss is a function of particle range and cell geometry. At a pressure of 4 atmospheres of a 1:1 $F_2:N_2$ mixture the average range of fission fragments is 0.5 cm. Using this and the cell dimensions itemized above, a loss of 10% is estimated by Steinberg's analysis of energy deposition (18).

A more serious difficulty associated with dose calculations is the decomposition of uranium hexafluoride. In experiments utilizing 0.1 g of ^{235}U about 30% of the UF_6 was not recovered. This amount was nearly the same for 1-hr and 7-hr exposures, which indicates that the UF_6 reaches an equilibrium with its solid decomposition product in an hour or less. A 5-fold change in neutron flux had little effect on the percentage of UF_6 recovered. The decomposition of UF_6 was subject to the $N_2:F_2$ ratio, the extent increasing as the fluorine content decreased. In addition, the percentage of UF_6 recovered was less in those experiments using lesser amounts of UF_6 .

In making corrections for this phenomenon the following assumptions were made: (a) the reaction between UF_6 and its solid decomposition product reaches equilibrium within a few minutes, (b) the solid product forms a thin, uniform coating on the walls of the aluminum cells, and (c) the fission events occurring in the solid deposit 50% of their energy in the gas phase and the remainder in the walls of the cell. The energy losses calculated on these premises were 10 to 20% for those experiments with 0.1 g of ^{235}U and a $N_2:F_2$ ratio of 1:1.

The aluminum cells for the fission fragment studies were used for only a single experiment with one exception. One cell was used for three experiments in the earlier work. This cell was not cleaned between runs, and some uranium remained on the walls due to decomposition of UF_6 . This fact was taken into account in interpreting the data.

RESULTS AND DISCUSSION

The N-F compounds identified as products of these synthesis studies were nitrogen trifluoride, cis-difluorodiazine and trans-difluorodiazine. Mass spectra were the primary means of identification but the first two compounds were also detected by infrared analysis. The retention times of the products in GC analysis were identical to those of NF_3 , cis- N_2F_2 , and trans- N_2F_2 . Tetrafluorohydrazine was not detected by gas chromatography. A small peak in the chromatogram close to that expected for N_2F_4 was always positive, whereas N_2F_4 gave a negative signal at the filament currents utilized in the thermal conductivity detector. Mass spectral analysis of this peak indicated it was sulfur hexafluoride. The NF_2 and NF mass peaks, the most abundant mass fragments for N_2F_4 (14), were quite small, and the N_2F_4 concentration was estimated to be no more than 0.0005% of the total sample.

Nitrogen trifluoride is the major product formed. The percentage yield of this compound is a linear function of the amount of energy deposited. Although one of the early experiments gave a low NF_3 concentration at a high dose rate,* the data plotted in Fig. 1 indicate the equilibrium concentration of NF_3 under these conditions is in excess of 1.7%. The 1-hr and 7-hr experiments gave data which can be plotted on the same curve. The G values for NF_3 formation at the 1:1 $N_2:F_2$ ratio were 0.25 to 0.44 molecules/100 ev. The 1-hr runs gave G_{NF_3} values which were in the lower part of this range.

*The aluminum cell for this experiment had been used in previous experiments and was not cleaned for this experiments.

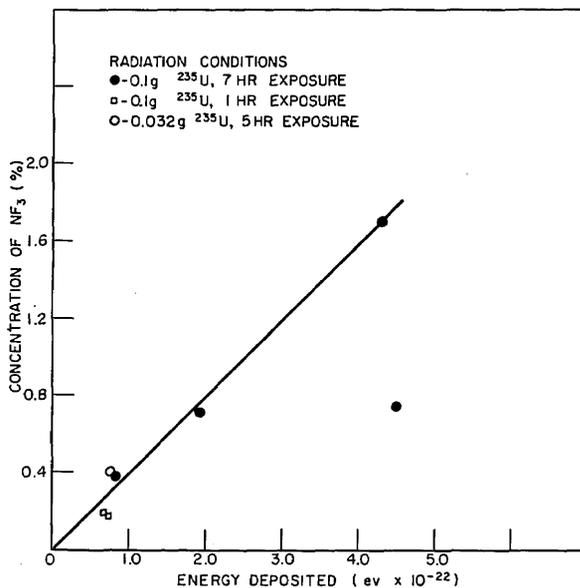


Fig. 1 - The NF_3 yield as a function of energy deposited

Cis-difluorodiazine is found in significant amounts, although the concentration of this product was never as high as that of NF_3 . It differs from NF_3 in that the amount formed is not a function of the total energy deposited. The percentage yield of cis- N_2F_2 decreases as the amount of energy deposited increases. In experiments using the same amounts of ^{235}U , N_2 , and F_2 , the amount of cis- N_2F_2 can be related to the thermal neutron flux. As shown in Fig. 2, the percent concentration of cis- N_2F_2 increases sharply as the thermal neutron flux decreases. At a thermal neutron flux of 1×10^{11} neutrons/cm²-sec, the amount of cis- N_2F_2 formed was the same for 1-hr and 7-hr experiments. The G values for cis- N_2F_2 production varied greatly, depending upon the neutron flux and the length of irradiation. The range observed was 0.01 to 0.27 molecule/100 ev.

The third N-F compound observed, trans-difluorodiazine, was found in minor amounts. In the majority of experiments it constituted 8 to 10% of the amount found for the cis compound, although the amount was as low as 3% and as high as 14%. This fairly constant percentage would indicate that these isomers are in equilibrium with each other at the time of formation.

The fluorine-to-nitrogen ratio has an effect upon the amounts of N-F compounds produced. This phenomenon, which was explored to a limited extent, is complicated by the fact that the quantity of UF_6 retained in the gas phase is dependent upon the $\text{N}_2:\text{F}_2$ ratio. As a consequence the calculation of energy deposited in mixtures with a low $\text{F}_2:\text{N}_2$ ratio are more subject to error. Thus, the experimental results with the 1:3 fluorine-to-nitrogen ratio are considered less reliable than the other results shown in Table 1.

The 1:1 ratio affords the highest yield of NF_3 . The yield is decreased to a greater extent by a low nitrogen content than by a low fluorine content. The change in ratio, at comparable neutron fluxes, appears to alter the cis- N_2F_2 G value very little. When the differences in thermal neutron flux are considered, it is noted that the ratio is important. For instance, the concentrations for both the 1:3 and 3:1 $\text{F}_2:\text{N}_2$ ratios fall above the curve in Fig. 2 for cis- N_2F_2 concentration based on the 1:1 ratio. A high fluorine content is particularly favorable from the viewpoint of the thermal neutron flux.

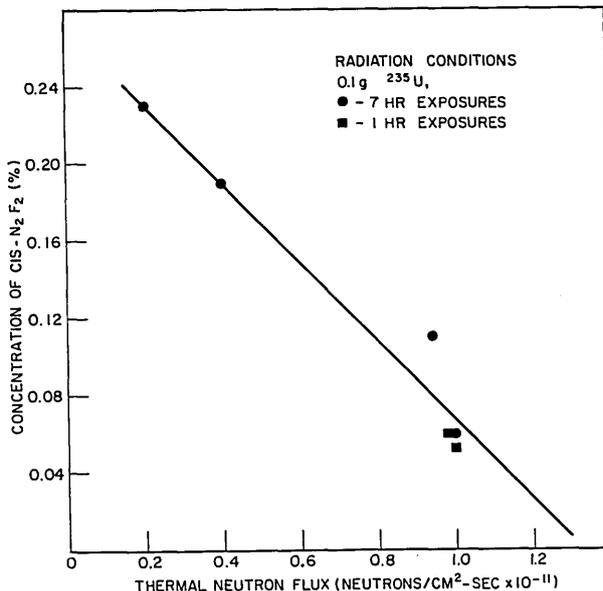


Fig. 2 - The cis-N₂F₂ yield as a function of the thermal neutron flux

Table 1
Effect of the F₂:N₂ Ratio on Yield

F ₂ :N ₂ Ratio	Thermal Neutron Flux (neutrons/cm ² -sec)	NF ₃ Yield		cis-N ₂ F ₂ Yield	
		Conc. (%)	G Values (molecules/100 ev)	Conc. (%)	G Values (molecules/100 ev)
1:1	0.94 × 10 ¹¹	1.70	0.40	0.11	0.026
3:1	1.23 × 10 ¹¹	0.84	0.14	0.17	0.028
1:3	1.17 × 10 ¹¹	1.10	0.30	0.10	0.027

The formation of NF₃ by bombardment of a nitrogen-fluorine mixture with fission fragments has been reported by Aerojet-General Nucleonics (9). The work detailed here shows that the system is more complicated than that report indicated, since two other compounds, cis- and trans-difluorodiazine, are significant products. The G_{NF₃} for the Aerojet-General Nucleonics experiment was estimated to be 0.33 to 0.67 molecule/100 ev. This value is in the same range as the values found in this work (0.25 to 0.44 molecule/100 ev), but the uncertainties in energy deposition for the Aerojet-General Nucleonics experiment precludes any reliable comparisons.

An interesting aspect of this investigation is the marked difference in involvement of the two major products, NF₃ and cis-N₂F₂. The formation of NF₃ increases with the total energy input, and the equilibrium concentration of this compound is greater than 1.7%, the highest concentration attained. For cis-N₂F₂ the dose rate is more important than the total dose.

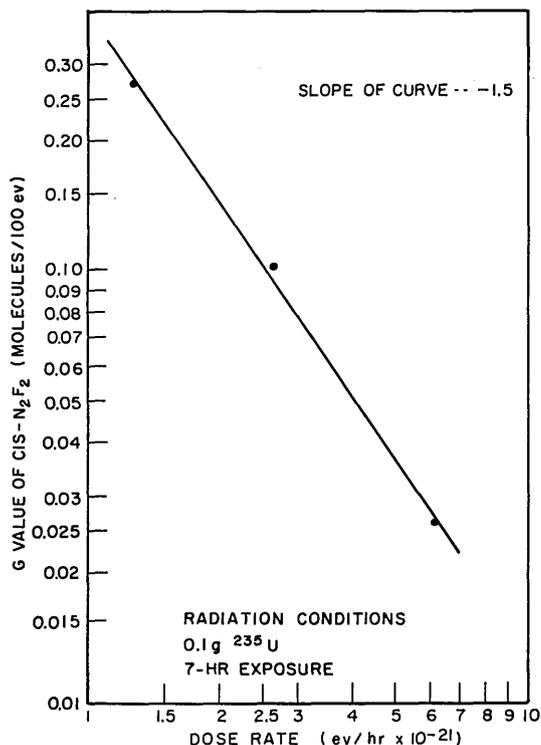


Fig. 3 - The cis-N₂F₂ yield as a function of the dose rate

The variation of yield with dose rate has been observed in other radiation studies. Ozone formation from oxygen under the influence of alpha-rays (19) and beta-rays (20) decreased as the dose rate increased. Radiation studies with gamma rays (21) and mixed pile nuclear radiation (22) also revealed the dose rate is a controlling factor in the alkylation of unsaturated hydrocarbons. The alkylation studies indicated the G values to be inversely proportional to the square root of the dose rate, a fact which was considered indicative of a radical-radical termination step of a chain reaction. On a theoretical basis, Mund (23) predicted that the G value for ozone formation should also be inversely proportional to the square root of the radiation intensity.

The influence of the dose rate on the yield of cis-N₂F₂ is much greater than in the cases cited above. This is evident from the log-log plot of $G_{\text{cis-N}_2\text{F}_2}$ versus dose rate. The slope of this curve in Fig. 3 is -1.5 indicating

$$G_{\text{cis-N}_2\text{F}_2} = \frac{K}{(\text{dose rate})^{3/2}}$$

The fact that the same concentration of cis-N₂F₂ is obtained for radiations of 1 hr and 7 hr indicates equilibrium between synthesis and decomposition can be attained under the conditions used. This was tested by an experiment in which 0.29% cis-N₂F₂ was added to the initial reaction mixture. Exposure to a thermal neutron flux of 0.74×10^{11} neutrons/cm²-sec decreased the concentration to 0.12%. The datum point thus obtained would fall on the curve (Fig. 2) derived for experiments starting with no N₂F₂ present. The equilibrium thesis is corroborated.

A blank experiment, one without UF₆ added, was carried out to assess the contribution of pile radiations on formation of products. The total energy absorbed from the gamma-ray flux in this experiment was estimated to be 3% of the energy deposited in a comparable experiment with 0.1 g of ²³⁵U present. Approximately equal amounts of NF₃ and cis-N₂F₂

were formed. The small quantities found were about that to be expected for the gamma-ray flux from the reactor. This was shown by the radiation experiments on the $N_2:F_2$ system with gamma rays from a cobalt-60 source which are outlined below.

A second blank included 0.29% cis- N_2F_2 in the reaction mixture. The gamma-ray absorbed dose was of the same magnitude as in the other blank run. The cis- N_2F_2 concentration increased to 0.35% and a nitrogen trifluoride concentration of 0.024% was attained. Once again the amounts of these compounds produced were close to those expected on the basis of corollary studies with ^{60}Co radiation.

The radiation intensity can be varied by altering the neutron flux or the quantity of ^{235}U . The data shown in Fig. 3 were obtained by flux variation. An experiment in which the ^{235}U content was reduced to 1/3 the amount normally used gave complicating results. The quantity of NF_3 was that expected from the total absorbed dose, but the cis- N_2F_2 yield was only 40% of that expected on the basis of the dose rate found in Fig. 3. The amount of cis- N_2F_2 was somewhat closer (75%) to that predicted if the neutron flux were controlling (Fig. 2). It is concluded that a simple relationship between dose rate and cis- N_2F_2 yield cannot explain all of the phenomena observed in the described experiments.

The results of the ^{60}Co radiations referred to above are interesting to compare with the chemonuclear results. The major product is cis- N_2F_2 and the G value for this compound is independent of gamma-ray dose rate for the range studied, 0.04×10^{19} to 4.9×10^{19} ev/hr. The G value of 0.6 molecule/100 ev is greater than that found for any of the chemonuclear experiments. At the highest concentration attained (0.33%) there was no evidence of approach to equilibrium.

The yield of NF_3 was less for gamma-ray synthesis than with chemonuclear synthesis. G_{NF_3} was 0.22 molecule/100 ev at a dose rate of 4.9×10^{19} ev/hr and 0.23 molecule/100 ev at a dose rate of 1.0×10^{19} ev/hr. The G value dropped to 0.09 molecule/100 ev at the lowest dose rate of 0.04×10^{19} ev/hr, but the analytical results were not precise because of the small amount of NF_3 produced in this experiment. The only other N-F compound identified in the gamma-ray syntheses was trans- N_2F_2 . This material was in equilibrium with cis- N_2F_2 as the amount found was always 7 to 10% of the amount of the cis isomer.

A mechanism for the reaction between nitrogen and fluorine in a radiation field cannot be suggested on the basis of the limited data available from this study. It is evident, however, that this reaction is a complicated one, since it is dependent upon the linear energy transfer (LET) and the radiation dose rate. It is interesting to note that the conditions which favor formation of the less stable compound N_2F_2 (low LET and low dose rate), are those which give lower concentrations of radicals and ions on a localized scale and on a gross scale. The effect of LET may not be real, however. Additional experiments with fission fragments at dose rates intermediate between the gamma-ray studies and the fission fragment studies reported here would determine the importance of LET vs dose rate for N_2F_2 production. If the $G_{N_2F_2}$ yield reached a limiting value of 0.6 molecule/100 ev at intermediate dose rates, the LET would be shown to be unimportant for N_2F_2 formation. The data indicate that LET is a factor for NF_3 formation, since the G value, which varied little with fission fragment dose rate or with gamma-ray dose rate, was higher for fission fragment radiation than gamma radiation.

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APPENDIX

MEASUREMENT OF THERMAL NEUTRON FLUX

The radiations were performed in the pool of the NRL research reactor. An aluminum stand which fitted into the core grid plate held the radiation cell in one of 26 positions. The flux was measured initially at low reactor power with indium foils at different positions on the aluminum stand. These measurements, which were carried out by Reactor Branch personnel, gave a linear plot for distance from core vs log neutron flux.

As the program progressed, these measurements were inadequate for two reasons: The core loading of the reactor must be changed periodically to maintain the desired power level, and the cell could not be precisely aligned. The neutron flux would vary due to each of these factors. Consequently the flux was monitored for each radiation.

Gold foils 0.001 inch thick were used in these measurements. The use of bare and cadmium-covered foils permitted the calculation of the activity due to thermal and fast neutron fluxes. The cadmium covers were 0.015 inch thick in keeping with the considerations used in previous flux determinations.*

The NRL calibrated graphite pile with Ra-Be source was used for standardization purposes.* One to two week exposures of 2.5-cm² foils were made with this low flux source. Reactor experiments used foils approximately 0.02 cm² in area because of the high flux level. The foil weights were determined on a microbalance to the nearest microgram.

To avoid foil geometry difficulties in comparing the standards with the unknowns, the foils were put into solution for counting. Each foil was dissolved in the minimum amount of aqua regia necessary for complete solution. Following evaporation to dryness, potassium cyanide solution was added to the test tube to form a stable cyanide complex with the gold. The pile radiated foils were dissolved, evaporated, and redissolved in the test tubes used in the counting. An aliquot of the solution from the reactor exposed foils was used. In each case the volume of liquid counted was 3.0 ml. The KCN concentration was 1N for the small foils, but 2N KCN was necessary to dissolve the large foils from the calibration exposures. The counting error due to this difference in concentration was 0.5%, an amount which could be accepted for these measurements.

The counting was performed in a well-type scintillation counter. The counting efficiency (counting rate for standard divided by the calculated disintegration rate) was found to be 0.387 and 0.393 for two different calibrations, which is satisfactory agreement for these studies.

The ratio of activity of the cadmium covered foils to the activity of the bare gold foils was less than 0.1 in all cases and varied but slightly from one reactor position to another. This is illustrated in Table A1 for two thermal neutron flux levels. The ratio was higher on the side of the cell toward the reactor core (front) than in the other positions. The finding that the ratio of cadmium covered foil to bare foil activity varied little with flux level and cell position permitted the use of only bare foils in the experiments which followed. The activity of the bare foil in such a case was reduced by the proper amount (8%) to determine the activity due to the thermal neutron flux.

*Bebbs, E. H., and Price, H. C., Jr., "Naval Research Laboratory Research Reactor, Part VII - Neutron Flux Measurements and Power Determination," NRL Report 5196, Nov. 1958.

Table A1
Gold Foil Activity Due To Thermal and Fast Neutrons

Thermal Neutron Flux (neutrons/cm ² -sec)	Ratio of Cadmium Covered Foil to Bare Foil Activity at Various Positions on the Cell			
	Front	Back	Left Side	Right Side
0.94×10^{11}	0.090	0.072	0.072	0.081
0.204×10^{11}	0.092	0.082	0.084	0.069

The results of the measurements of the thermal neutron flux in four positions on the exterior of the aluminum cell are listed in Table A2. Four foils were heat sealed in a 0.004-inch-thick polyethylene sleeve such that each foil was displaced 90 degrees around the outside diameter of the cell. The flux in the position closest to the reactor (front) was the highest and the flux in the back the lowest as expected. The flux in the left position was normally lower than that in the right position. This is because the radiation cell was to the left of the center of the reactor core. The average of the thermal neutron fluxes in the four positions was used in calculating the energy deposited in the cell contents.

Table A2
Thermal Neutron Flux For Fission Fragment Experiments

Experiment	Thermal Neutron Flux (neutrons/cm ² -sec $\times 10^{-11}$)				
	Front Position	Back Position	Left Side	Right Side	Average
6	1.07	0.85	0.88	0.95	0.94
7	0.232	0.186	0.194	0.203	0.204
8	1.33	1.07	1.14	1.14	1.17
9	1.37	1.14	1.17	1.22	1.23
11	0.91	0.75	0.77	0.79	0.80
12	0.80	0.64	0.67	0.68	0.70
13	0.84	0.68	0.71	0.72	0.74

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13. ABSTRACT Mixtures of nitrogen and fluorine containing uranium-235 as uranium hexa-fluoride have been exposed to the radiation field of a nuclear reactor. The fission fragments formed by reaction of ^{235}U with the thermal neutron component of the reactor radiation have caused the formation of nitrogen fluorides. Those identified were nitrogen trifluoride, cis-difluorodiazine, and trans-difluorodiazine. Nitrogen trifluoride is the major product, and the amount formed is dependent upon the total energy deposited. The other two products are in equilibrium with each other, and the amount formed decreases as the radiation intensity increases. The total G value for compound formation is less than 1 molecule per 100 electron volts.			

14. KEY WORDS	LINK A		LINK B		LINK C	
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Chemical reactions Synthesis (chemistry) Nitrogen fluorides Nuclear reactors Fission products Rocket propellants Thermal neutrons Uranium hexafluoride Radiation chemistry						

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