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Modeling of the $H + NF_2$ Reactive Flow

J. M. HERBELIN, M. A. KWOK, and N. COHEN
Laboratory Operations ✓
The Aerospace Corporation
El Segundo, Calif. 90245

15 April 1981

Interim Report

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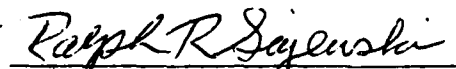
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This interim report was submitted by The Aerospace Corporation, El Segundo, CA 90245, under Contract No. F04701-80-C-0081 with the Space Division, Deputy for Technology, P. O. Box 92960, Worldway Postal Center, Los Angeles, CA 90009. It was reviewed and approved for The Aerospace Corporation by W. R. Warren, Jr., Director, Aerophysics Laboratory. Major Ralph R. Gajewski, SD/YLXT was the project officer for the Mission Oriented Investigation and Experimentation Program.

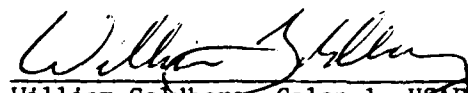
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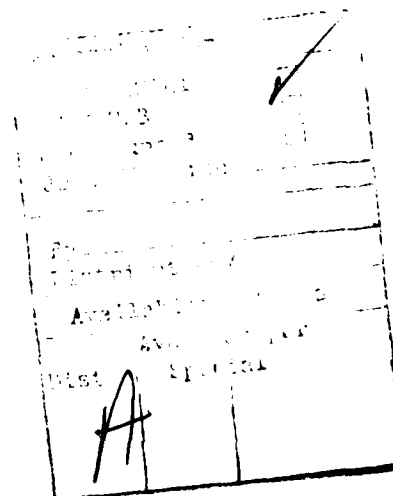
of these two species along the flow distance. This agreement indicates a value for the rate coefficient for $H + NF_2 \rightarrow HF + NF(a^1\Delta)$ of $2.0 \times 10^{13} \text{ cm}^3/\text{mol-sec}$; for the transfer $HF(3) + NF(a^1\Delta) \rightarrow HF(1) + NF(b^1\Sigma)$, a value of $6.0 \times 10^{13} \text{ cm}^3/\text{mol-sec}$ at room temperature is indicated. The analysis also provides an indirect determination of the radiative lifetime of the $NF(a^1\Delta)$ between 0.6 and 1 sec.

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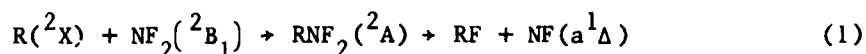
FIGURES

1.	Observed Number Densities as a Function of Flow Tube Axial Position Z.....	5
2.	Radial Variations of Selected Species at Z = 10 cm.....	7
3.	Model Fit of $H + NF_2$ Production of $NF(a^1\Delta)$ Showing Sensitivity of Rate Constant.....	9
4.	Model Fit of $NF(b^1\Sigma^+)$ Density vs. Axial Position Z.....	10
5.	$NF(a^1\Delta)$ Density vs. Axial Position Z for a Condition When [D] << $[NF_2]$	14
6.	Peak $NF(a^1\Delta)$ Density as a Function of NF_2 Flow Rate.....	15

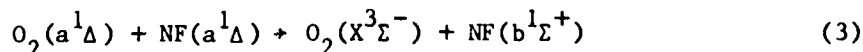
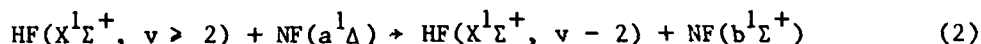


MODELING OF THE H + NF₂ REACTIVE FLOW

Herbelin and Cohen¹ proposed that an addition-elimination rather than an abstraction mechanism might be the preferred reaction channel for the production of electronically excited NF(a¹Δ) from the ground-state reagents R(= CH₃, H or D) and NF₂(²B₁):



It was found that the electronically excited NF(b¹Σ⁺), which is responsible for the green (5288 Å) emission, can be produced from the NF(a¹Δ), either by a vibration-electronic energy-transfer process [reaction(2)] involving HF(v > 2) or an electronic-electronic energy-transfer process [reaction(3)] using O₂(a¹Δ).



Recently, Kwok, Herbelin, and Cohen² measured the cross sections for the removal of NF(b¹Σ⁺) by HF(v = 0), H and D atoms, and the radiative lifetime for the NF(b¹Σ⁺) state. They reported values for the rate coefficients for

¹J. M. Herbelin and N. Cohen, "The Chemical Production of Electronically Excited States in the H/NF₂ System," Chem. Phys. Lett. **20** (6), (1973), p. 605.

²M. A. Kwok, J. M. Herbelin, and N. Cohen, "Collisional Quenching and Radiative Decay Studies of NF(a¹Δ) and NF(b¹Σ⁺)," in Electronic Transition Lasers, J. I. Steinfeld, ed., MIT Press, Cambridge, Massachusetts (1976), pp. 8-12.

the deactivation of $\text{NF}(a^1\Delta)$ by the above species, but were unable to make a direct determination of the radiative lifetime for this NF state. Since a knowledge of this radiative cross section is essential to the quantitative analysis of this system and to the determination of the system's potential as a chemical laser, we have attempted to extract this information from the previously reported³ NF density plots.

The number densities of HF and NF were observed as a function of axial distance on the large-diameter, fast-flow, medium-pressure flow tube facility.² Those experimental results are summarized in Fig. 1. Since a radiative decay rate of $\text{NF}(a^1\Delta)$ has not been experimentally established, we report photon flux instead of density alone.

Our starting point in analysis was a kinetic model developed in the Aerophysics Laboratory that closely approximates the observed $\text{HF}(X^1\Sigma^+, v = 1, 2, 3)$ vibrational levels as a function of the flow distance.⁴ Also shown in Fig. 1 is a comparison of the theoretical and experimental HF density plots⁴ for a standard flow case. The initial conditions for this computation are as follows:

$$[\text{F}] = 6.0 \times 10^{-11} \text{ mol/cm}^3$$

$$[\text{H}_2] = 5.9 \times 10^{-10} \text{ mol/cm}^3$$

$$[\text{Ar}] = 5.3 \times 10^{-8} \text{ mol/cm}^3$$

³M. A. Kwok, J. M. Herbelin, and N. Cohen, Collisional Quenching and Radiative Decay Studies of $\text{NF}(A^1\Delta)$ and $\text{NF}(b^1\Sigma^+)$, TR-0077(2940)-6, The Aerospace Corporation, El Segundo, California (15 April 1977), p. 16.

⁴M. A. Kwok and R. L. Wilkins, private communication. This is an earlier version of model discussed in R. L. Wilkins and M. A. Kwok, "Temperature Dependence of $\text{HF}(V_1 = 1) + \text{HF}(v_2 = 0)$ Vibrational Relaxation," J. Chem. Phys. **70**, 1705 (1979).

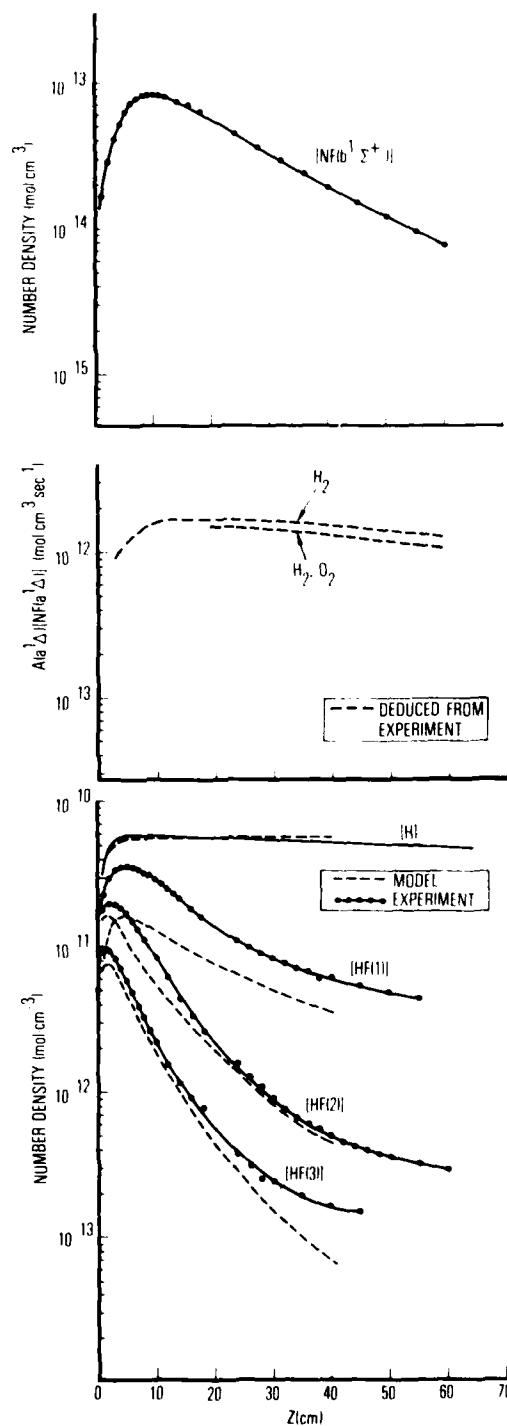


Fig. 1. Observed Number Densities as a Function of Flow Tube Axial Position Z

$$P = 1.0 \text{ Torr}$$

$$T = 298^\circ\text{K}$$

$$\bar{U} = 6.94 \times 10^3 \text{ cm/sec}$$

These conditions are determined experimentally. The empirical velocity $\bar{U}$ is 1.6 times the average velocity $\bar{U}$ determined from the continuity equation. In previous studies, this velocity has been found effective in establishing the characteristic time "scale" of kinetic phenomena.² The kinetic and flow model fits HF(v = 2) and HF(v = 3) and H densities well out to 30 cm. The HF(v = 1) is consistently predicted low, although the relative decaying spatial trend is duplicated. However, HF(v = 1) is not important in the following kinetics discussions.

A nonequilibrium chemistry and constant pressure flow computer code (NEST)⁵ was used to model the full cross section or radial elements of the axisymmetric cylindrical tube flow. A second code was used to integrate and average properties in the radial elements along the tube diameter while correcting for the paraboloidal velocity profile. Corrections for radial variations were found to be small. Peak HF(v = 2, 3) densities were altered less than 15% of values found by running the NEST code for the full cross section. Peak NF values were reduced <16%. In the regions of decay, all densities were adjusted slightly. In Fig. 2, typical computed radial variations for key species are shown at z = 10 cm for the final modeled case. We concluded from these studies that species were sufficiently uniform in cross section in the zone of NF(a¹Δ) and NF(b¹Σ⁺) production. In particular, previous observations² suggested that, once produced, NF(a¹Δ) should be insensitive to wall effects.

⁵E. B. Turner, G. Emanuel, and R. L. Wilkins, The NEST Chemistry Computer Program, TR-0059(6240-20)-1, The Aerospace Corporation, El Segundo, California (30 July 1970).

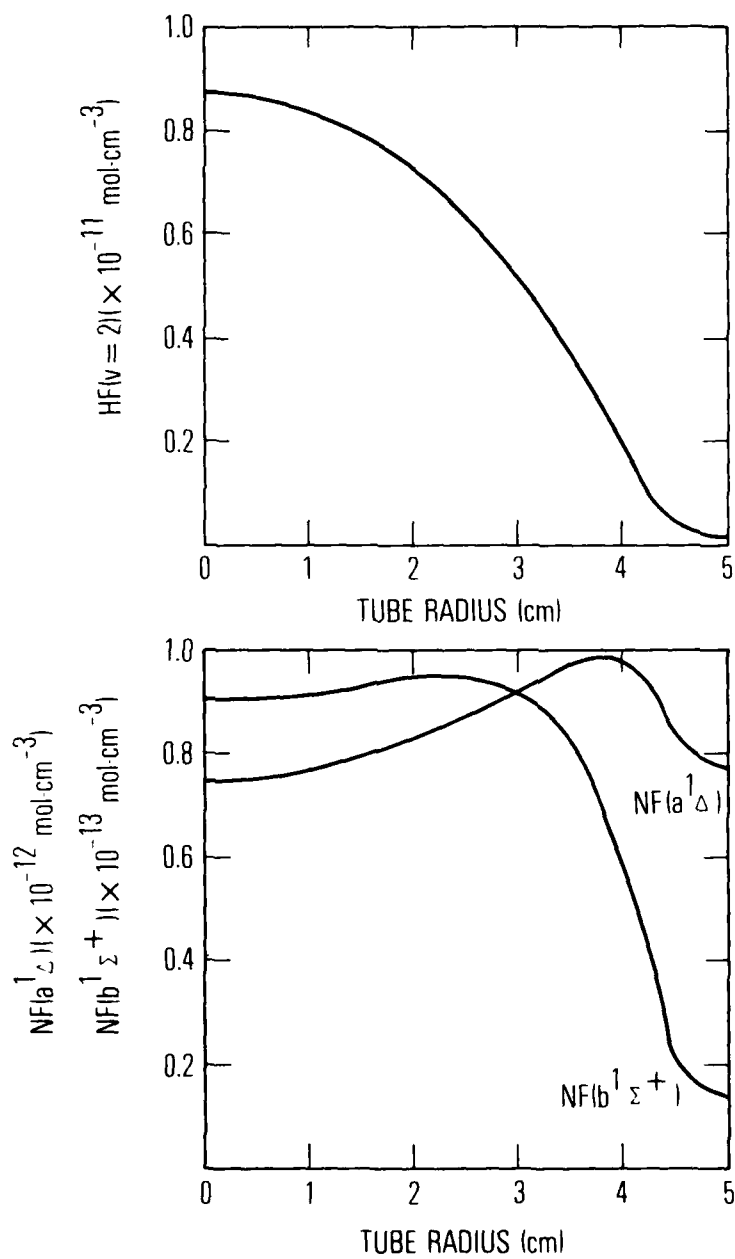


Fig. 2. Radial Variations of Selected Species at $Z = 10 \text{ cm}$

In Fig. 3, we attempt to fit the observed $A(a^1\Delta)$ $[NF(a^1\Delta)]$ results. The rate coefficient for reaction (1) with $R = H$ was estimated to be 2×10^{13} $\text{cm}^3/\text{mol-sec}$, based on its similarity to the $H + NO_2 \rightarrow OH(^2\Pi, v = 0) + NO$ reaction.⁶ Through the use of this value and the measured rate coefficient for the removal of $NF(a^1\Delta)$ by H atoms, we were able to reproduce the shape of the experimentally measured $[NF(a^1\Delta)]$ curve. We chose an initial value of $[NF_2] = 1.33 \times 10^{-12}$ mol/cm^3 for the case, but because the H atom concentration was excessive, the shape is insensitive to this concentration up to values of 10^{-11} mol/cm^3 . As can be readily seen from Fig. 3, however, the curve shapes are quite sensitive to the value of the rate coefficient for reaction (1). The original choice for $[NF_2]$ yields the best agreement with experiment.

We next introduced the transfer process (2) with a rate coefficient of $k_2 = 5 \times 10^{12}$ $\text{cm}^3/\text{mol-sec}$, calculated from the measured value for the reverse process, the deactivation of $NF(b^1\Sigma^+)$ by $HF(v = 0)$.² This implicitly assumes that electronic-vibrational energy transfer is the process involved. By using this value, we find that the resulting computed zone plots have shapes similar to the "A" curves in Fig. 4, with a peak value of $[NF(b^1\Sigma^+)] = 4 \times 10^{-14}$ mol/cm^3 at $z = 16.5$ cm downstream. This can be compared with the experimental value of 8×10^{-14} mol/cm^3 as shown in Fig. 4. By varying the initial $[NF_2]$, it was possible to vary the computed peak $[NF(a^1\Delta)]$ and $[NF(b^1\Sigma^+)]$ values over an order of magnitude range without appreciably affecting the shapes of the curves for NF , HF , or H .

The shape of the experimental curve for $NF(b^1\Sigma^+)$ relative to the computed curve "A" indicated that the following additional transfer process between HF and NF is also important:



⁶D. L. Baulch, D. D. Drysdale, D. G. Horne, and A. C. Lloyd, Evaluated Kinetic Data in High Temperature Reactions, 2, London; Butterworths (1973) p. 445.

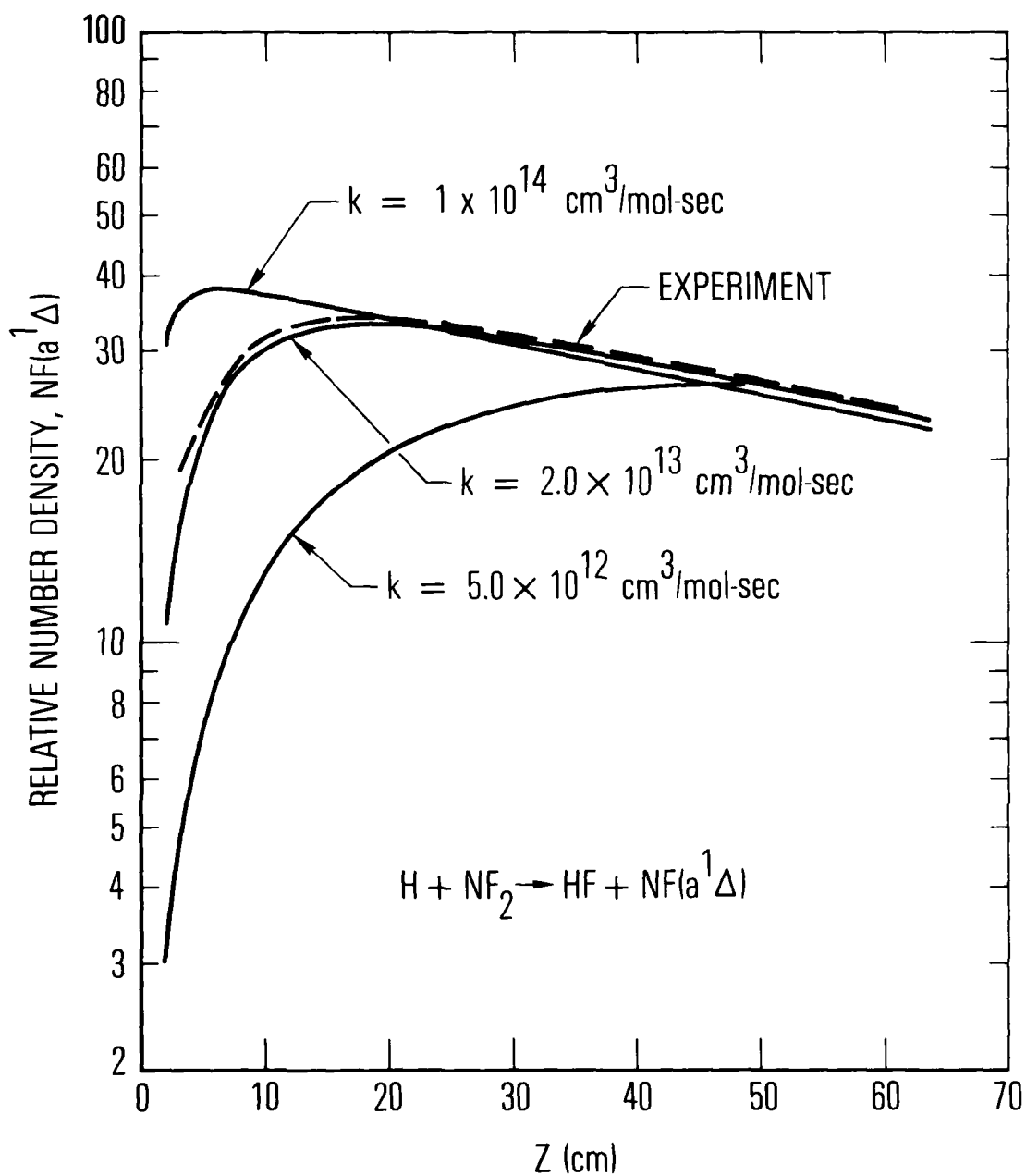


Fig. 3. Model Fit of $H + NF_2$ Production of $NF(a^1\Delta)$ Showing Sensitivity of Rate Constant

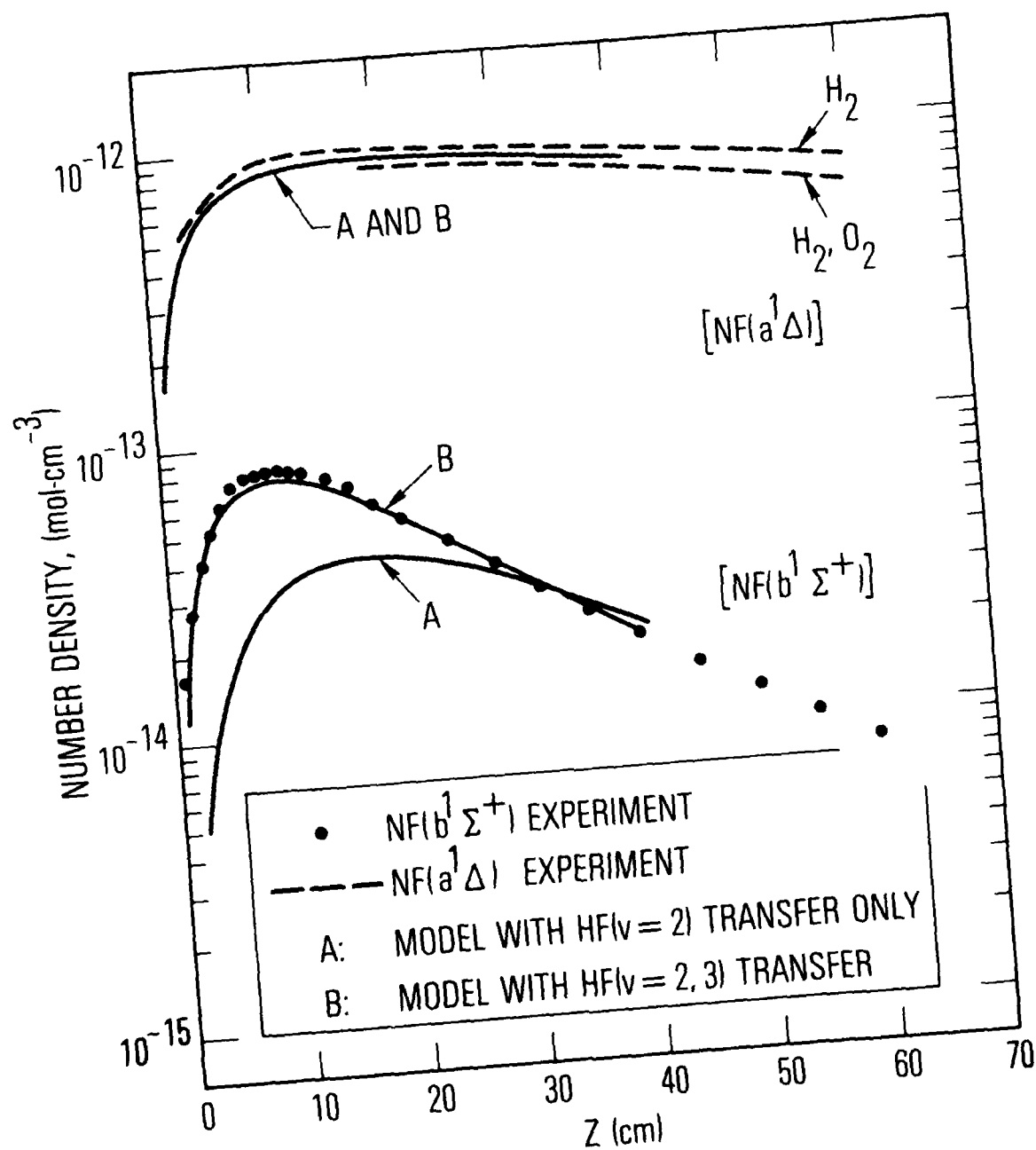
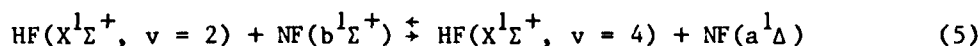


Fig. 4. Model Fit of $\text{NF}(b^1\Sigma^+)$ Density vs. Axial Position Z

Previous experience with other transfer processes between $\text{NF}(a^1\Delta)$ and HF , $\text{O}_2(a^1\Delta)$ and more recently I^* atoms⁷ indicates that energy resonance could be used to estimate a value for the rate coefficient for reaction (4). The exothermicity for process (2) is 298 cm^{-1} as written, assuming all species to be in the $J = 0$ rotational state. Transfer process (4) is 50 cm^{-1} exothermic. Since this is less than the 110 cm^{-1} for the transfer process between I atoms and NF , for which a gas kinetic cross section has been measured, we assumed that the vibrational energy of the $\text{HF}(v = 3)$ would transfer in one out of every two collisions. By using an approximate collisional diameter of $3 \text{ \AA}$ for all species involved, we arrived at a room temperature rate coefficient of $k_{(4)} = 6 \times 10^{13} \text{ cm}^3/\text{mol-sec}$ in the exothermic direction. The endothermic reverse reaction rate coefficient was then calculated to be $k_{(-4)} = 6 \times 10^{13} \exp(-142/RT)$.

Inclusion of this transfer process into the model resulted in considerably improved agreement between the shapes of the computed and measured curves. By trial and error, we adjusted this rate coefficient and the initial NF_2 concentration to the final values of $k_4 = 2.6 \times 10^{12} T^{1/2}$, $k_{-4} = 2.6 \times 10^{12} T^{1/2} \exp(-142/RT) \text{ cm}^3/\text{mol-sec}$, and $[\text{NF}_2] = 4.2 \times 10^{-13} \text{ mol/cm}^3$ to arrive at the agreement between the computed curves B and the experiment. Only a 50% adjustment from the initial estimate was required to achieve this excellent fit.

Calculations were also performed to explore the effect of the additional transfer process,



⁷J. M. Herbelin and M. A. Kwok, Electronic Angular Momentum Transfer in $\text{I}(^2P_{1/2}) + \text{NF}(a^1\Delta)$ System, TR-0076(6240-40)-4, The Aerospace Corporation, El Segundo, California (17 March 1976).

We found that the computed $NF(b^1\Sigma^+)$ profiles were quite insensitive to the process even when a gas kinetic rate coefficient was assumed. Therefore, no conclusion concerning this process is possible.

When this agreement between observed and computed values of $[NF(b^1\Sigma^+)]$ and its axial variation has been obtained, the computed population of the $NF(a^1\Delta)$ can be used to infer the radiative lifetime (inverse of the radiative decay rate) of the $NF(a^1\Delta) \rightarrow NF(X^3\Sigma^-)$ transition. We simply take the ratio of the observed integrated band intensities² from the $(b^1\Sigma^+)$ and $(a^1\Delta)$ states which is proportional to the ratio of wavelengths, populations, and radiative decay rates.

$$\frac{I(b^1\Sigma^+ \rightarrow X^3\Sigma^-)_{\text{expt1}}}{I(a^1\Delta \rightarrow X^3\Sigma^-)_{\text{expt1}}} = \frac{\lambda(a^1\Delta)}{\lambda(b^1\Sigma^+)} \frac{N(b^1\Sigma^+)}{N(a^1\Delta)} \frac{A(b^1\Sigma^+)}{A(a^1\Delta)} \quad (6)$$

We find that the ratio of radiative decay rates

$$\frac{A(a^1\Delta)}{A(b^1\Sigma^+)} = 0.015$$

which yields a value of $A(a^1\Delta) = 0.94 \text{ sec}^{-1}$ when the value of $A(b^1\Sigma^+) = 67 \text{ sec}^{-1}$ as measured by Kwok, *et al.*² is used. This method requires only a relative intensity calibration. The value for $A(a^1\Delta)$ was also computed independently on the basis of the absolute $(a-X)$ emission band intensity. Absolute intensities yield results such as the photon flux plot in Fig. 1. If $NF(a^1\Delta)$ is known, $A(a^1\Delta)$ is readily determined. In this case $A(a^1\Delta)$ is deduced to be 1.75 sec^{-1} . The two calculated values agree to within a factor of two.

As a final check of results from this analytical procedure, we performed an experiment in which we went from the hydrogen-rich regime, $[H] \gg [NF_2]$, to one in which the NF_2 was in slight excess. We accomplished this by decreasing the NF_3 flow and discharge power until we reach a fluorine atom flow, measured by our standard testing,² of approximately 0.05 of the test case. The total

pressure was maintained at the 1 Torr value and excess deuterium was used to produce a concentration of D atoms of 2.3×10^{-12} mol/cm³. We then introduced NF₂, produced by passing N₂F₄ through an oven prior to injection into the flow tube. The NF₂ was injected approximately 20 tube diameters (2 m) upstream of the N₂ injector to ensure adequate mixing. The NF₂ flow was calibrated by supplying an excess of N atoms from an additional microwave discharge through N₂ and then measuring by the standard method the F atoms produced by the reaction



The reduced concentration of the D atoms resulted in greatly extended NF(a¹Δ) zones, in which the concentrations of NF(a¹Δ) were approximately flat at distances in excess of 40 cm. Figure 5 shows the NF(a¹Δ) density plot for such a NF₂ flow condition. The peak is now reached much later in Z because the production rate $k_1[\text{NF}_2]$ is much slower than the previous $k_1[\text{H}]$ of the $[\text{H}] \gg [\text{NF}_2]$ regime.

In Fig. 6, we show the variation of [NF(a¹Δ)] deduced from the measured peak signals with the use of the above deduced 1.75 sec⁻¹ value for the radiative decay rate of NF(a¹Δ). Excess O₂ was added in one run to remove any interfering N₂ first positive emission, which is seen to have a 20% effect. Because of the limits of our N₂F₄ heating facility, we were not able to produce more than a 7.5 μmol/sec flow of NF₂. We see that the [NF(a¹Δ)] rises and appears to reach a limit of 3.0×10^{-12} mol-cm⁻³. This value is in the mid-range of limits of H atom density. The lower H limit was determined by an F-atom titration and by the assumption of full conversion to H atoms by the F + H₂ reaction. The upper H limit was given by the assumed total conversion to F by the NF₃ passed through the discharge. It appears that reaction (1) is the key mechanism in producing NF(a¹Δ) since its maximum density equals that of the limiting reagent H within a factor of two.

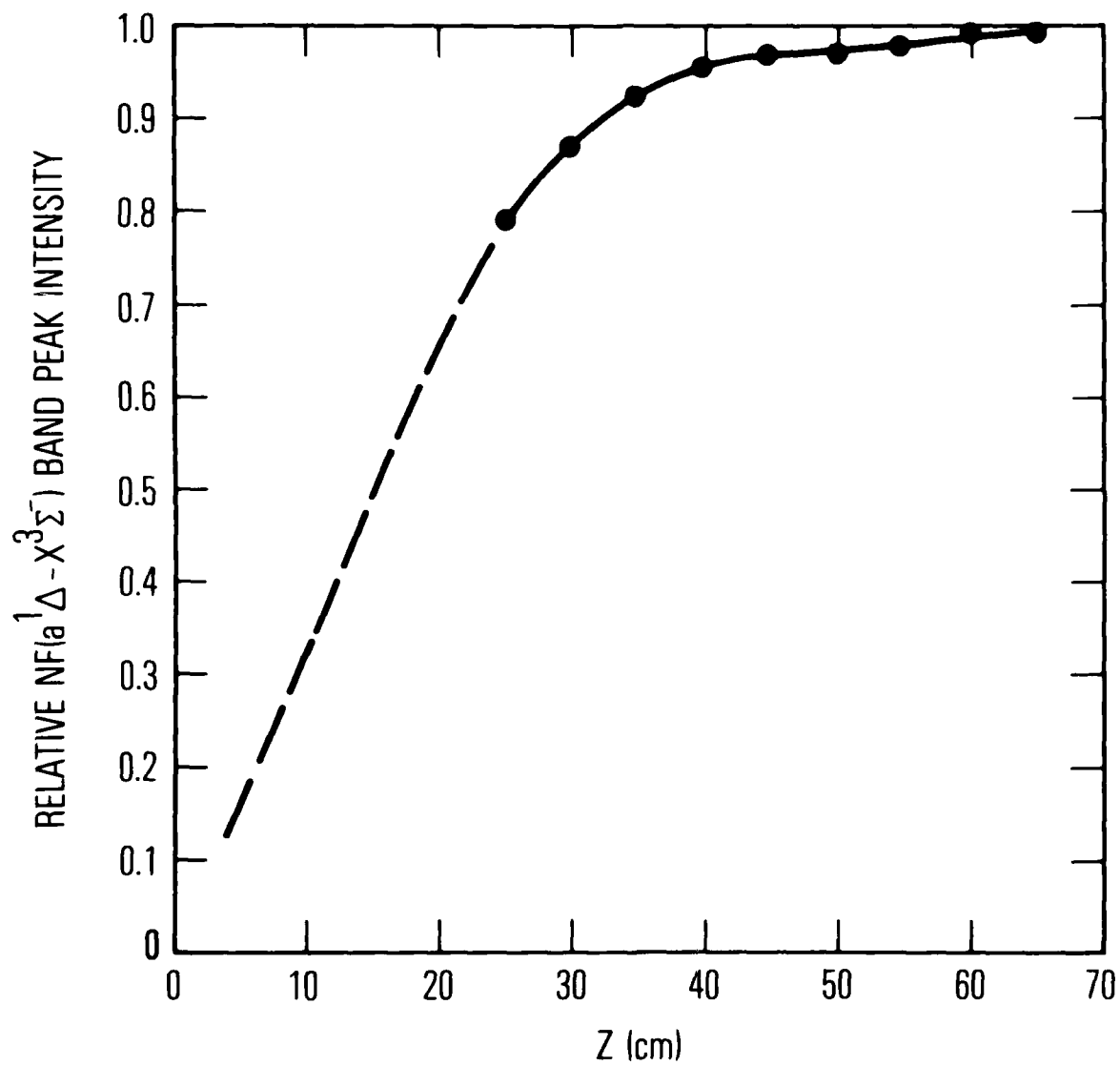


Fig. 5. $\text{NF}(a^1\Delta)$ Density vs. Axial Position Z For a Condition When $[D] \ll [\text{NF}_2]$

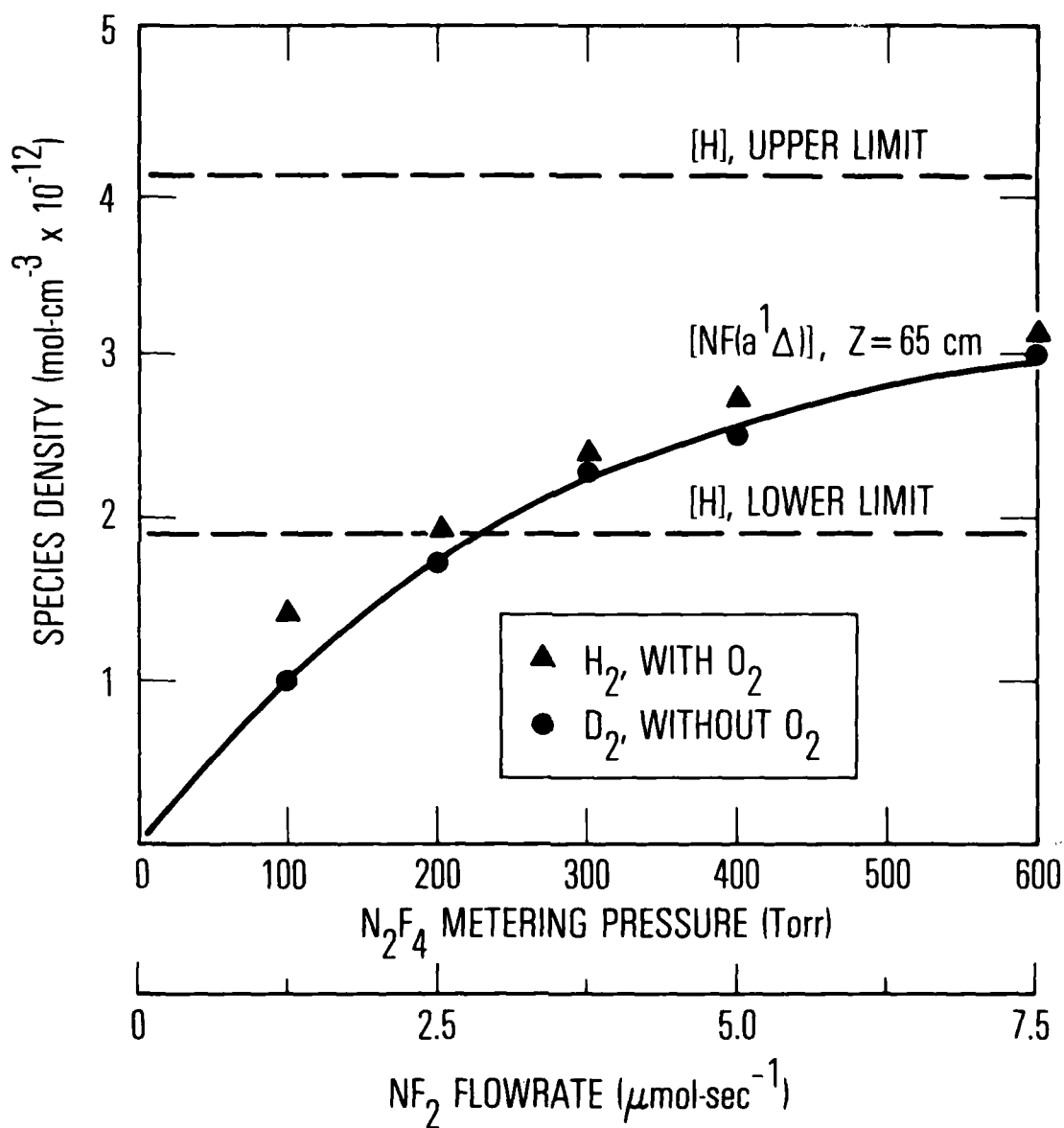


Fig. 6. Peak NF(a¹Δ) Density as a Function of NF₂ Flow Rate

This result supports the above analyses as well as the contention that reaction (1) is close to 100% efficient in producing electronically excited $\text{NF}(a^1\Delta)$. Further work is required to confirm this latter conclusion.

APPENDIX

Since this analytical work was done, independent experimental work has been reported by Cheah, Clyne, and Whitefield,⁸ and by Malins and Setzer.⁹ The experimentally determined rate coefficients (at room temperature) for reaction (1) can be compared with our analytical deduction in the following tabulation:

$10^{-13} k_1 (\text{cm}^3\text{-mol}^{-1}\text{sec}^{-1})$	
This work	2
Cheah, Clyne, Whitefield	0.90
Malins, Setzer	>0.37

We consider our deduction to be in good agreement with the measurement of Cheah, et al., which relied on direct observation by UV resonance absorption of fluorescence to monitor the disappearance of H atoms caused by the presence of NF_2 radicals. They did not resolve the problem of whether product state $\text{NF}(a^1\Delta)$ is in a totally inverted condition. If this assumption is made, then Cheah, et al. have succeeded in determining the reaction rate coefficient of reaction (1).

The measurements of Malins and Setzer confirm the exciting prospect that over 90% of the $\text{H} + \text{NF}_2$ reaction produces the excited electronic state, $\text{NF}(a^1\Delta)$. However, their rate measurement depends on a method of comparison

⁸C. T. Cheah, M. A. A. Clyne, and P. D. Whitefield, "Reactions Forming Electronically-excited Free Radicals," J. Chem. Soc. Faraday II **76** (1980) pp. 711-728.

⁹R. J. Malins and D. W. Setzer, "Rate Constant and Branching Ratio for Formation of Electronically Excited NF by Reaction of H Atoms with NF by Reaction of H Atoms with NF_2 ." (To be published). We thank Dr. Malins for advance preprints of his work and for many helpful discussions.

with another "baseline" reaction, and it may not be surprising that their absolute numbers are in only fair agreement with Cheah, et al. and this work. Their method of diagnosis prevents them from observing all the product states of HF(v), namely HF(v = 0), which is expected to be a significant product. Therefore, they indicate their rate coefficient could be larger.

Malins and Setzer have also attempted the first measurement of the NF(a¹Δ) radiative lifetime with a result of 5 to 6 sec. Our deduced results yielded 0.6 to 1 sec by two different approaches. We estimate the Malins number to be more reliable, but the factor of 5 disagreement suggests that even more experimental work should be done on this interesting reaction system.

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