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# CHEMICALLY-PRODUCED N<sub>2</sub>(A) TO NO(X) ENERGY TRANSFER IN A SUPERSONIC FLOW

Y. D. Jones

November 1987



Final Report

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Air Force Systems Command  
Kirtland Air Force Base, NM 87117-6008

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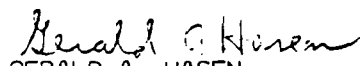
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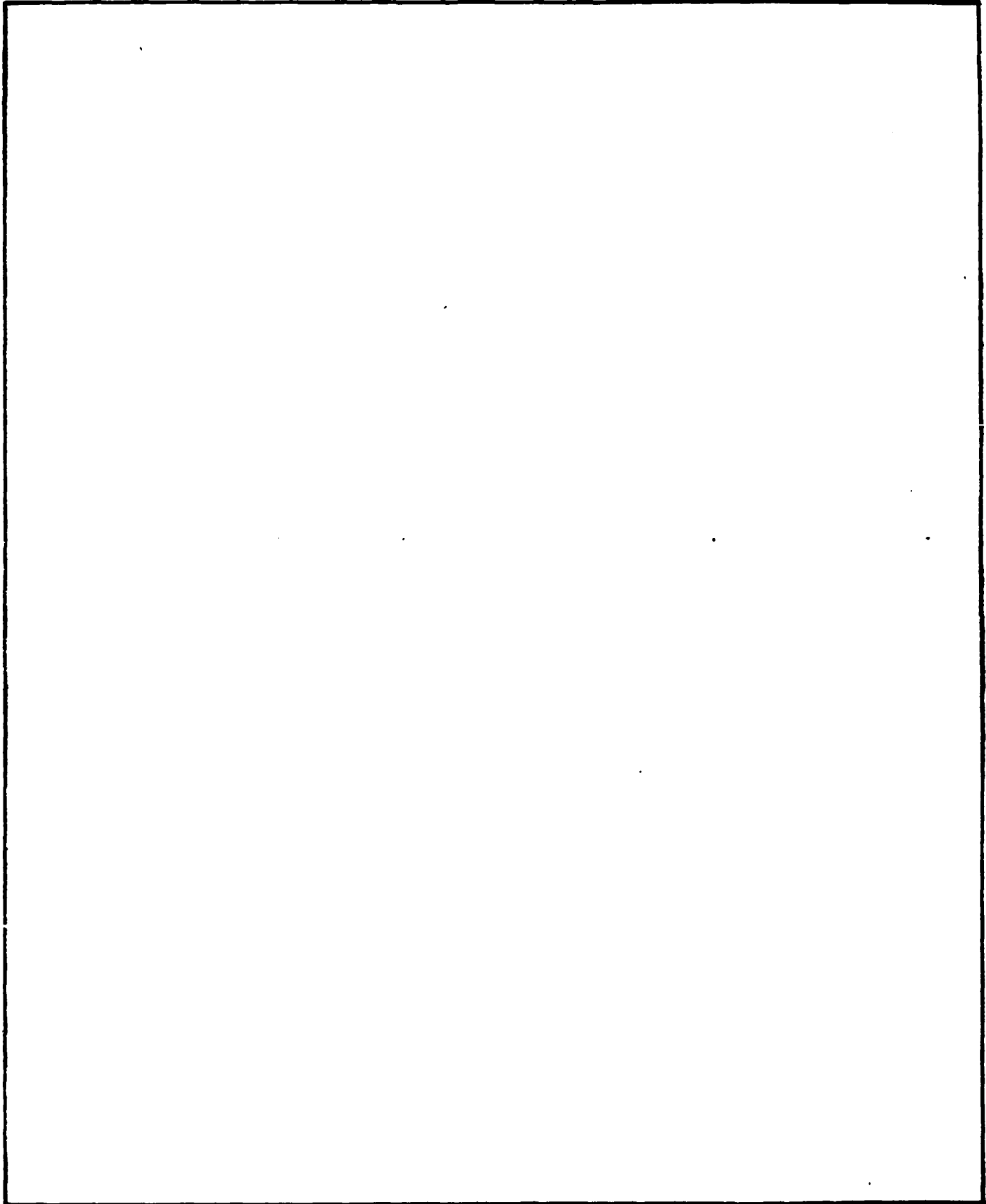
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PREFACE

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## INTRODUCTION

The  $N_2F_4 + H_2$  scheme for production of  $NF(a^1\Delta)$ ,  $NF(b^1\Sigma)$  and  $N_2(A^3\Sigma_u^+)$  is well-known from flow tube studies at low pressure and low temperature (Refs. 1-4). The  $N_2(A)$  is a good energy storage molecule with 6.2 eV of energy and a 2.0 s lifetime (Ref. 5). Because of the long lifetime of the species,  $N_2(A)$  does not make a good laser candidate. Transfer of the energy in the  $N_2(A)$  molecule to NO is well-documented (Refs. 6-8). The transfer is efficient, but has been demonstrated only in small-scale systems such as flow tubes and with nonchemically produced  $N_2(A)$ . This set of experiments was designed to demonstrate efficient NO excitation by  $N_2(A)$  in a purely chemical system as produced via the following reactions:



Although, the  $N_2(A)$  to NO transfer is efficient, when NO is added to the excited nitrogen system, the kinetics become complicated. A basic set of the  $N_2(A) + NO$  reactions and rates is given in the Appendix. The transition of interest in NO is the  $A \rightarrow X$ . The  $NO(A^2\Sigma)$  state has a lifetime of  $2.0 \times 10^{-7}$  s (Ref. 9). The Franck-Condon factor for the  $NO(A^2\Sigma, v' = 0)$  to  $NO(X^2\Pi, v' = 1)$  transition is the largest at 0.26 (Ref. 10). The  $NO(A-X)$  spectrum is complex with 8 branch bands (Refs. 11 and 12). The A-state rotational constant is

1.9965 and the Boltzmann rotational distribution is centered about  $J = 7$  (Ref. 13). The stimulated emission cross section can be calculated as  $1.03 \times 10^{-16} \text{ cm}^2$ . The NO molecule is a promising laser candidate and has been lased by optical pumping (Ref. 14).

Early work by Setser (Ref. 15) and Collear (Ref. 16) showed that the  $\text{N}_2(\text{A}^3\Sigma, v' = 1)$  to  $\text{NO}(\text{X}^2\Pi, v'' = 0)$  transfer resulted in  $\text{NO}(\text{A}^2\Sigma)$  in the ratio of  $v = 1$  to  $v = 0$  of 1:10 (Ref. 15) and 1:2 (Ref. 16). This is the dominant process since  $\text{N}_2(\text{A})$  in  $v > 1$  rapidly relaxes by V-V processes with  $\text{N}_2(\text{X})$  (Ref. 15). The most desirable  $\text{NO}(\text{A-X})$  transitions for lasing are the strong emission lines of  $v' = 0$  to  $v'' = 1, 2$ .

## DEVICE DESCRIPTION

## OVERVIEW

The overall system consisted of a 316L stainless steel chamber with viewing ports on four sides and has been previously described (Ref. 17). Figure 1 shows a top view of the chamber with positions shown for the gas input plumbing. The chamber was exhausted into a cooled diffuser in the transition section and two heat exchangers. The device was evacuated using two Kinney 850 cfm pumps with two M & D Pneumatics 2700 cfm blowers for a system total of 7,100 cfm.

The BCL - 16 nozzle was positioned in the chamber wall with the gas inputs. The BCL - 16 nozzle cross section is shown in Figure 2. The BCL - 16 nozzle was developed for HF/DF laser application (Ref. 18) and studied for those same systems (Ref. 19). For the  $N_2F_4 + H_2$  system the combustor portion of the assembly nozzle was operated as it had been designed to produce F atoms. The hydrogen or deuterium and fluorine were injected into the combustor along with helium diluent at a molar ratio of  $F_2:D_2:He$  of approximately 1:2:50.

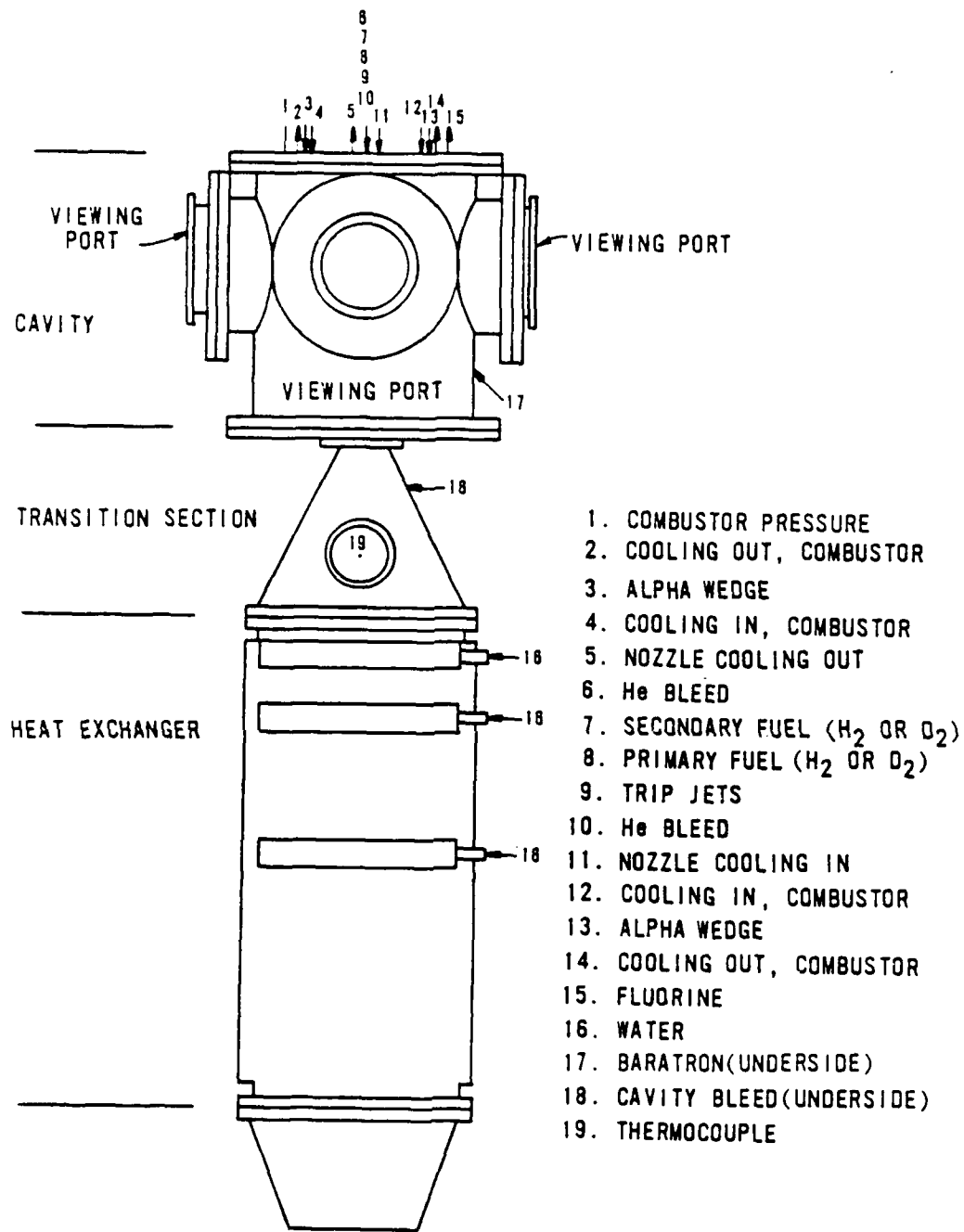


Figure 1. System overview.

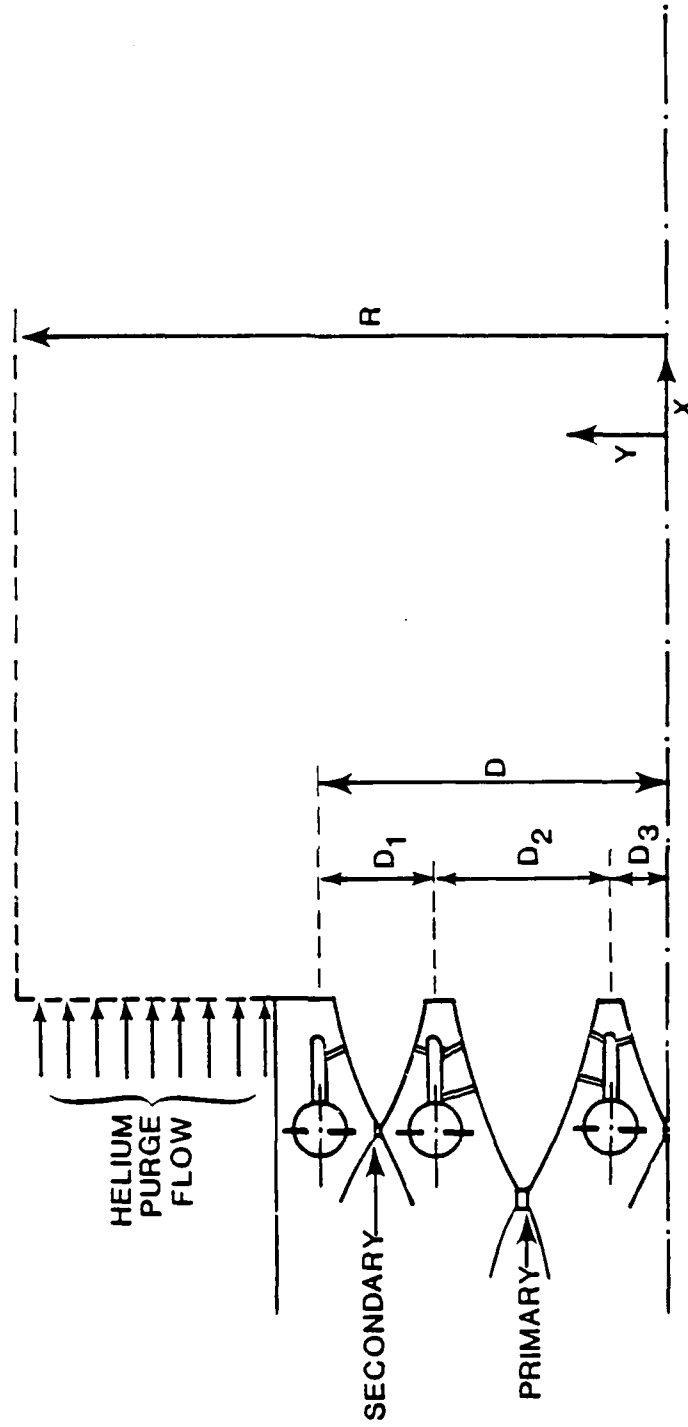


Figure 2. Cross section of the BCL-16 nozzle.

## NO INJECTOR SYSTEM

The NO was delivered to the system via a hypersonic wedge constructed from aluminum (Fig. 3). The wedge was used for preliminary examination of the NO injection. The gas was fed to the wedge by two tubes which also served as supports. The tubing could be moved along the centerline ( $X_C$ ) of the cavity. This allowed for optimization of the NO(A-X) emission by varying the injection position and the NO flow rate. The NO flow system is depicted in Fig. 4. The  $N_2$  or He could be provided through the purge system. During testing only He was used as a diluent. The entire system was operated remotely. For safety, however, an NO detector (Ecolyzer, Model 412) was placed near the device.

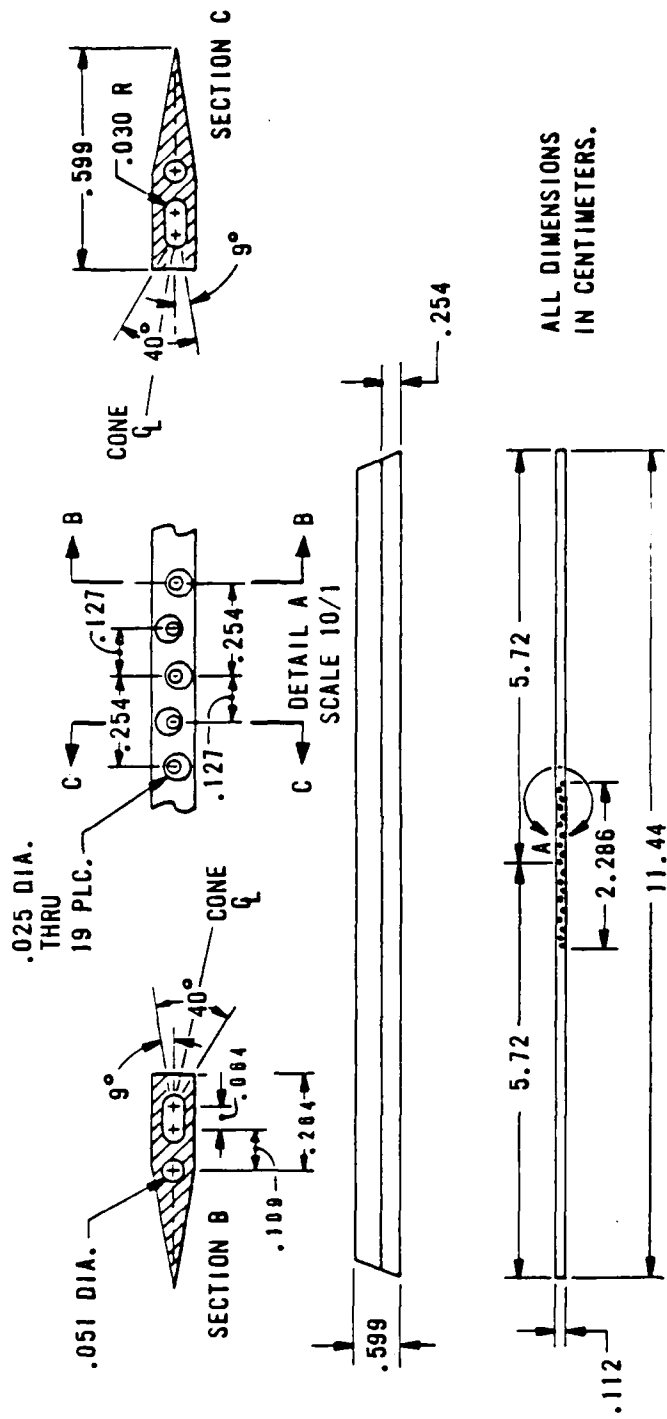


Figure 3. Detail of the NO injector.

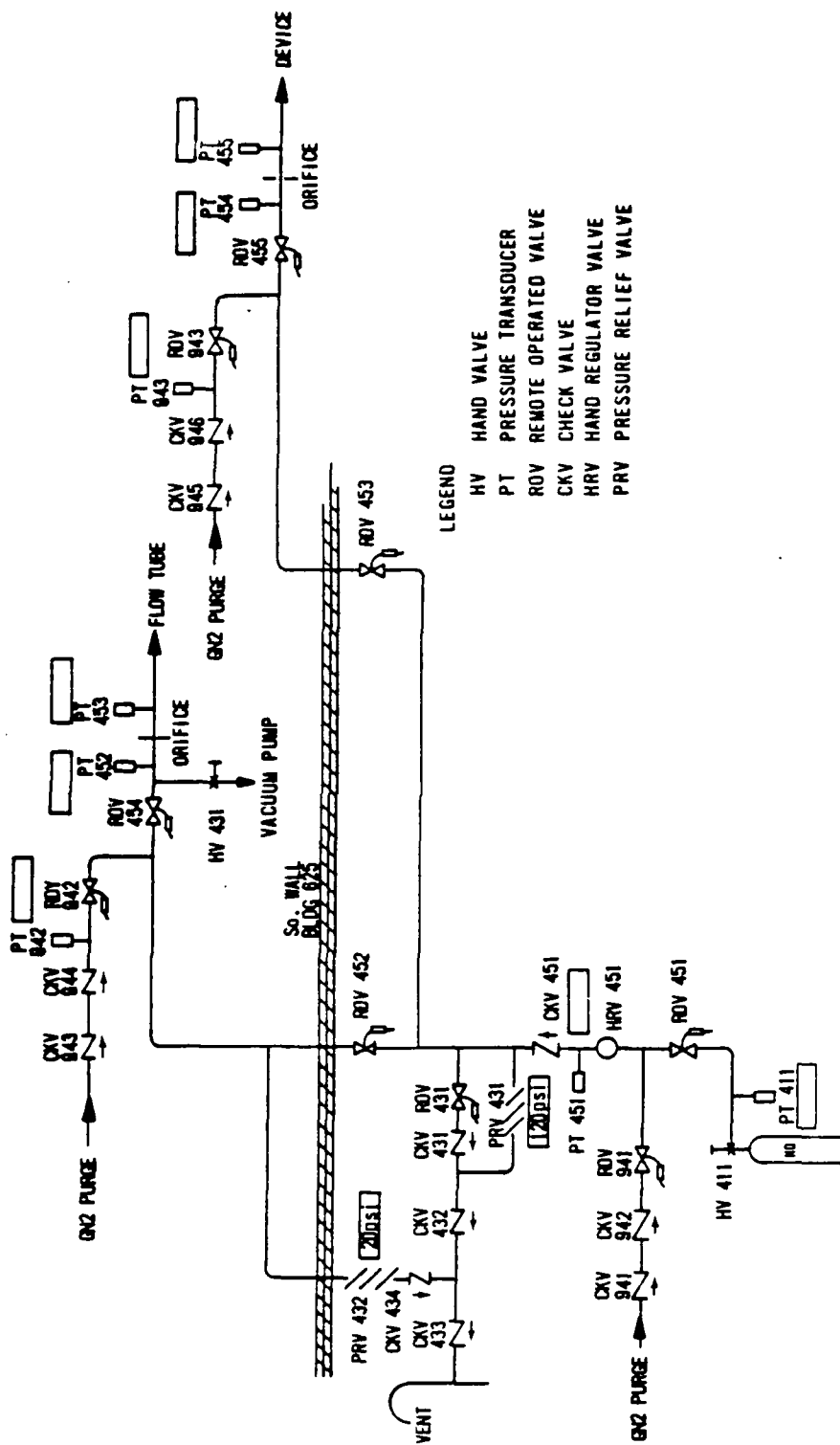


Figure 4. NO flow system.

## DIAGNOSTICS

NF( $a^1\Delta$ ) AND NF( $b^1\Sigma$ ) DIAGNOSTICS

The NF( $a^1\Delta$ ) diagnostic was an important part of the reaction analysis. The NF(a) and NF(b) diagnostics have been described (Ref. 20). The 874.2 nm emission from the NF(a-x) transition was detected via a 38.1 cm long spatial filter with 0.17 cm dia orifices coupled to a fused silica fiber optic. The fiber optic was bifurcated so that one end was fed into the NF(b) diagnostic. This allowed detection of NF(a) and NF(b) to be made within the same viewing volume. The diagnostic as applied to the device is shown in Fig. 5. The actual width of the flame was used to determine the volume viewed by the diagnostic. The NF(a) emission was filtered using an extremely narrow bandpass filter (FWHM 0.98 nm) centered at 874.29 nm which essentially eliminated interferences from the close-lying  $N_2(B)$  and HF( $v=3$ ) emissions.

The NF(b) diagnostic used the same bifurcated fiber optic with the output of the other porting of the cable going to a narrow bandpass filter centered at 531.4 nm and FWHM of 9.8 nm. The 538.8 nm emission of the NF ( $b^1\Sigma-x^3\Sigma$ ) transition was not masked by any near-lying emissions. The spatial filter was mounted on a remotely operated translation stage with a linear voltage displacement transducer (LVDT) to accomplish scans across the flow field of the device with a known position. Sample scans of the NF( $a^1\Delta$ ) and NF( $b^1\Sigma$ ) emissions are shown in Figs. 6 and 7.

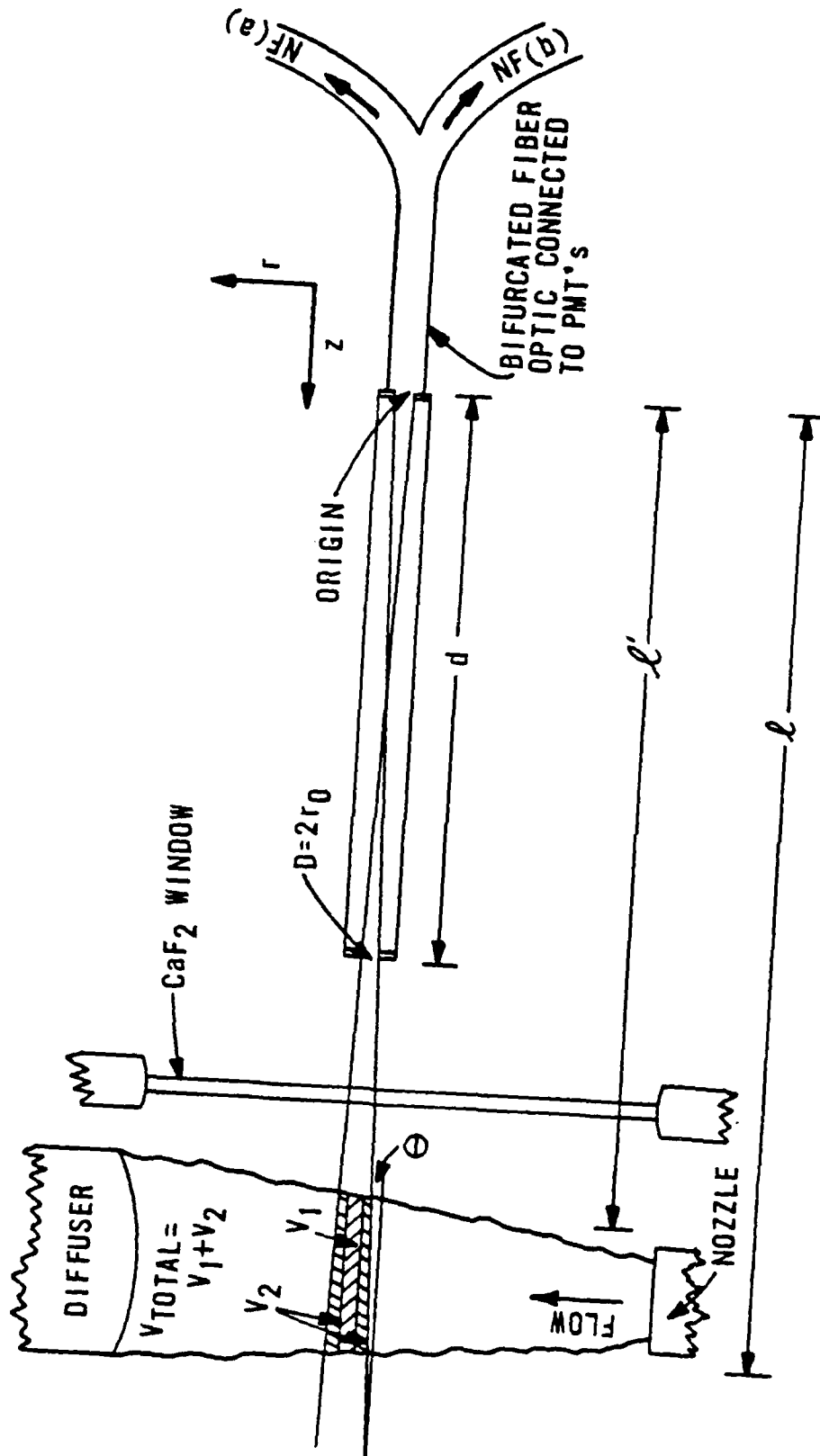


Figure 5. Diagnostic set-up.

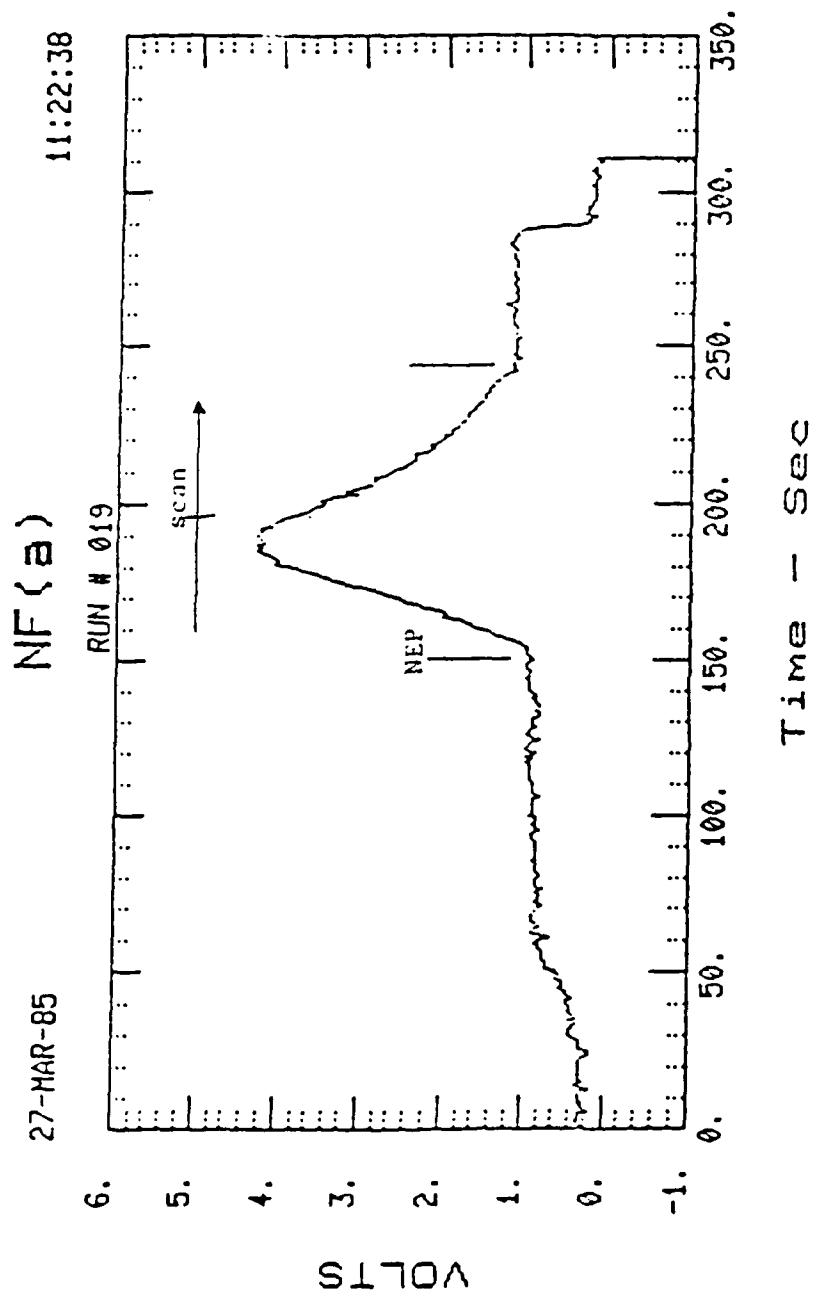


Figure 6. Sample NF(a) scan.

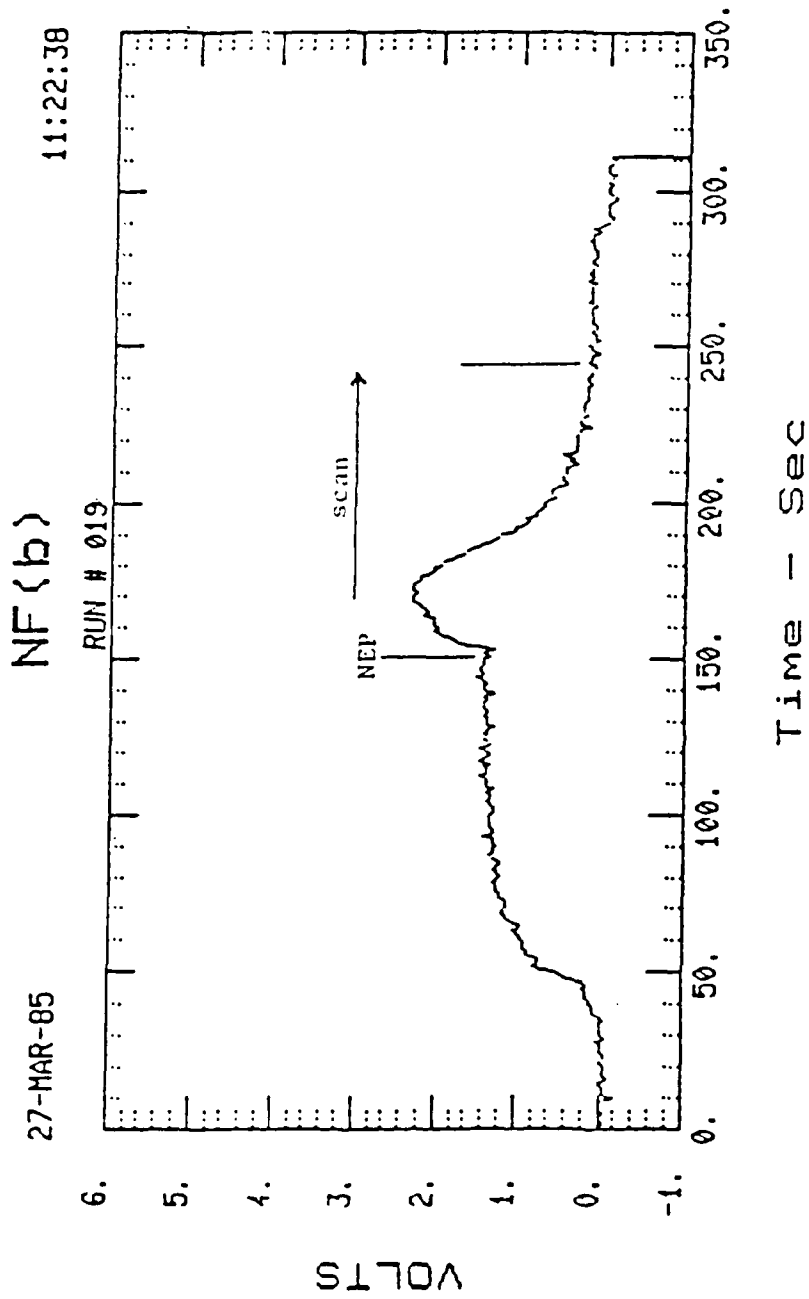


Figure 7. Sample NF(b) scan.

OPTICAL MULTICHANNEL ANALYZER (OMA)

The OMA III 1460R system (EG&G PAR) was used to monitor the change in emission over a wide wavelength range (usually 300-900 nm) at a fixed point within the device. The OMA III system consisted of a nonintensified diode array head (Model 1412) coupled to a Model 1233 polychromator.

N<sub>2</sub>(A) TO NO TRANSFER STUDIES

The system N<sub>2</sub>(A) production was optimized and has been described previously (Ref. 21). Maximum N<sub>2</sub>(A) production was found to occur when D<sub>2</sub> was used in place of H<sub>2</sub> due to better penetration of the higher molecular weight gas and fewer loss mechanisms for the NF(a<sup>1</sup>Δ) (Ref. 22). The two effects cannot, at this time, be decoupled because of only preliminary flow studies. The N<sub>2</sub>(A) was monitored by the visible N<sub>2</sub>(B) emission in the flow. The N<sub>2</sub>(B) relaxes to N<sub>2</sub>(A) with the emission of a photon. The N<sub>2</sub>(A) may also be produced directly. Therefore, the N<sub>2</sub>(b) gave a lower limit to the actual N<sub>2</sub>(A) production. The additional N<sub>2</sub>(A) contribution was evaluated by examination of the N<sub>2</sub>(c) population in previous experiments (Ref. 23). The N<sub>2</sub>(b) was determined to be accurate to within a factor of 10. Without the hypersonic wedge in place, maximum N<sub>2</sub>(B) was about  $2 \times 10^{11}$  molecules/cm<sup>3</sup>. The wedge interfered slightly with the flow field and, thus, mixing of the NF<sub>2</sub> and H<sub>2</sub> streams. The maximum N<sub>2</sub>(B) detected with the wedge in place was about  $1 \times 10^{11}$  molecules/cm<sup>3</sup>. Variation of the injection point along the X<sub>c</sub> was first performed. Figure 8 shows the variation of NO(A-X) emission (or γ-bands) with wedge position. The distance measured was from the backside of the wedge which had a width of 0.599 cm. The maximum NO(A-X) emission was at 1 to 2 cm from the nozzle exit plane (NEP).

Flow rate variation is shown in Figs. 9 and 10. Figure 9 shows the NO(A-X) emission as a function of flow rate of NO. The NO emission decreased with increased NO flow rate. This is probably due to disturbance of the mixing of primary, secondary and trip jets by large flow rate injection. The flow rate range was limited by the flow control system. The NO can react with N(2D) rapidly (Ref. 23),  $k = 7.0 \times 10^{11}$  cm<sup>3</sup>/molecules-s, providing a competitive pathway for N<sub>2</sub>(A) formation. There is a definite decrease in N<sub>2</sub>(B) with increasing NO flow rate (Fig. 10). The decrease is most probably due to a combination of these effects in addition to self-quenching. The effect of He diluent is demonstrated by the data on the graph in Fig. 9 at 0.181 g/s NO. The He effectively dilutes the flow and does not aid mixing.

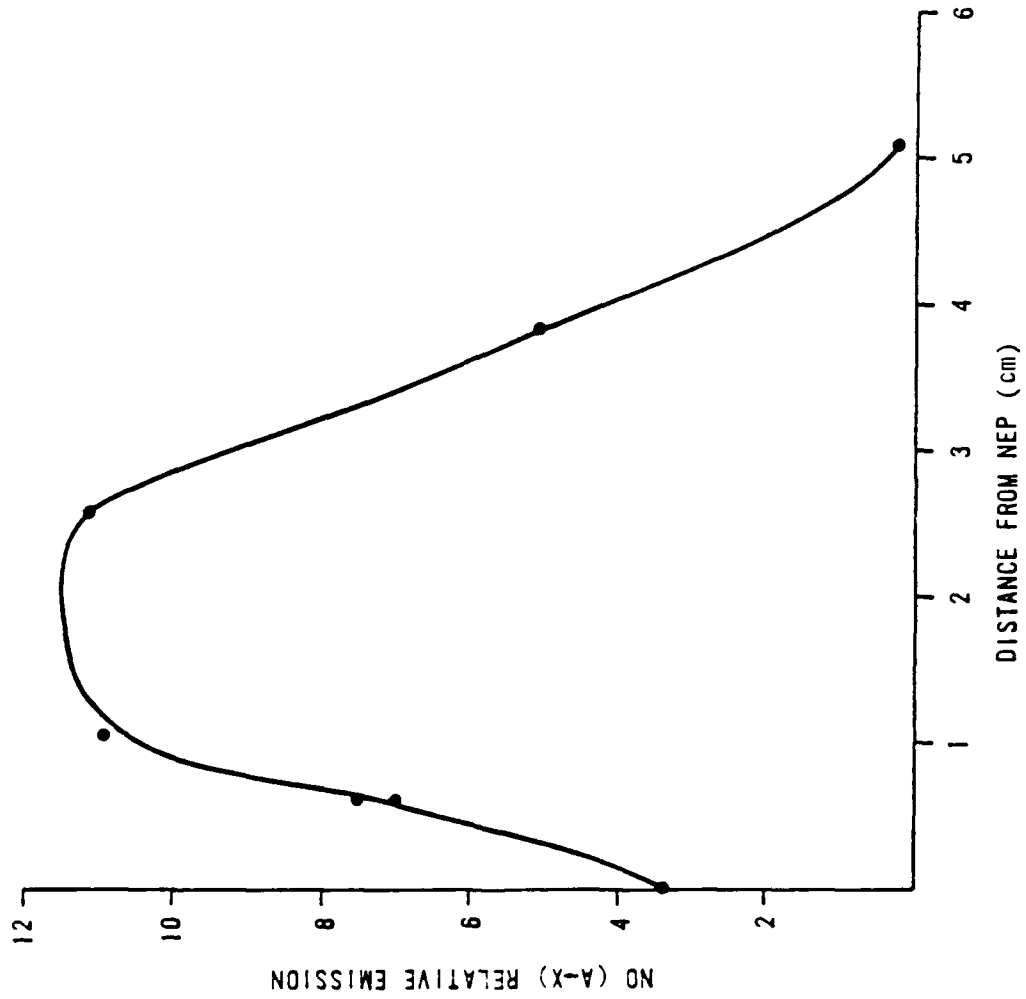


Figure 8. NO emission with variation in injector position.

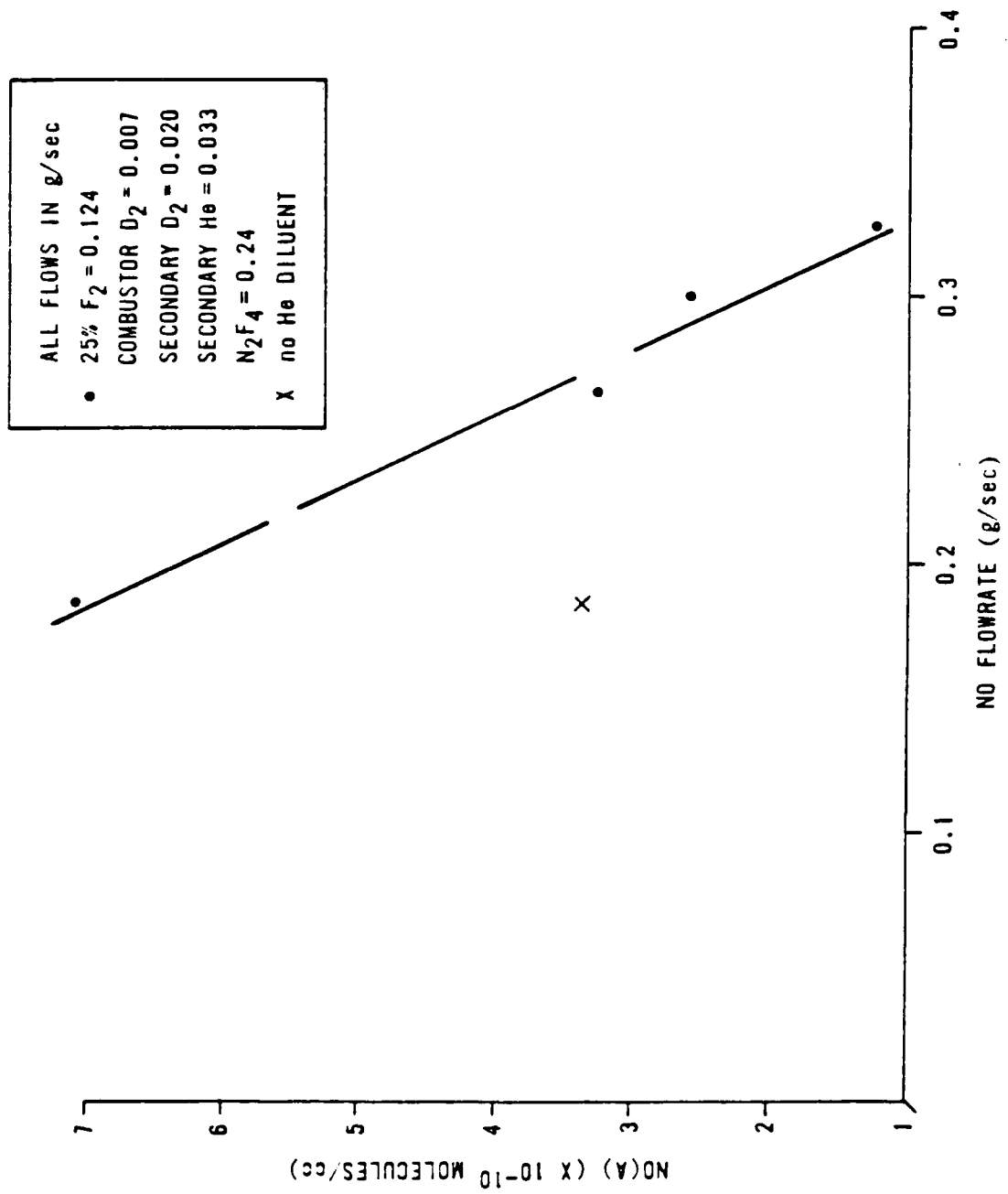


Figure 9. NO(A) emission as a function of NO flow rate.

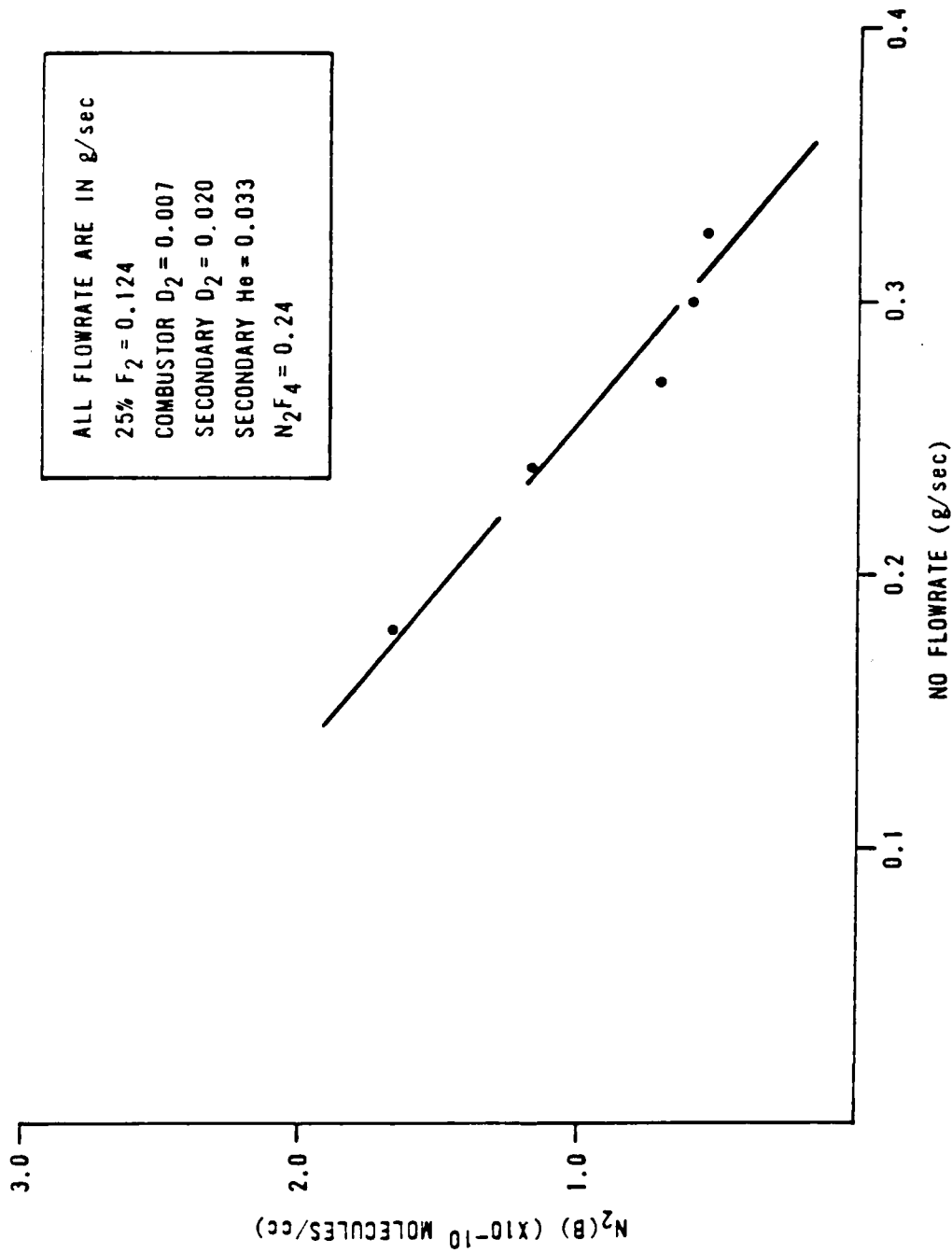


Figure 10. Variation of N<sub>2</sub>(B) emission as a function of NO flow.

A minimum number of runs were performed with trip jet injection of the  $D_2$  and secondary injection of the  $N_2F_4$ . These were designed to determine if mixing could be improved in this configuration, providing greater number densities of  $NO(A)$ . Table 1 contains information from the tests. There is some increase observed in the reversed configuration. The  $NO$  does not appear to interfere with  $NF(a^1\Delta)$  or  $NF(b^1\Sigma)$  production significantly as evidenced by the high concentrations in the tests.

Table 2 contains information from several tests where data was taken with and without  $NO$  flow. For the two high  $NO$  flow rate conditions, the efficiency is 30 to 50 percent due to the creation of turbulence in the flow. At the low flow rate, using the  $N_2(B)$  emission yields a greater than 100 percent transfer. What is actually occurring is that not all of the  $N_2(A)$  is accounted for by the  $N_2(B)$  emission. This also implies that the  $NF_2 + D_2$  reaction sequence forms some  $N_2(A)$  directly instead of all  $N_2(B)$  which then relaxes to  $N_2(A)$ . This is possible since the  $N_2(A)$  is not detectable directly in this system because of interferences from  $NO$  emission from contaminants in the  $N_2F_4$ .

A typical OMA III spectra with  $NO$  is shown in Fig. 11. The  $NO(A-X, v' = 0)$  and  $NO(A-X, v' = 1)$  peaks were identified. Specifically, the (0-1), (0-2), (0-3), (0-4), (0-5), (0-6), (1-6), (1-7) and (1-8) peaks were detected. Possible  $\beta$ -band peaks are also identified in the figure. Figure 12 was taken on the same test with the  $NO$  flow turned off and is included for comparison. The OMA III scans shown are not corrected for wavelength response of the detector.

TABLE 1. D<sub>2</sub> Trip Jet Injection.

Wedge	Primary		Trip	Secondary		(Molecules/cm <sup>3</sup> )			
	25% F <sub>2</sub> in He <sup>a</sup>	Combustor D <sub>2</sub> <sup>a</sup>		He <sup>a</sup>	NF <sub>4</sub> <sup>a</sup>	N <sub>2</sub> (B) X10 <sup>10</sup>	NO(A) X10 <sup>10</sup>	NF(a <sup>1</sup> Δ) X10 <sup>15</sup>	NF(b <sup>1</sup> Σ) X10 <sup>12</sup>
0.20	0.14	0.006	0.02	0.12	0.33	0.40	negl.	6.1	4.10
0.22	0.13	0.006	0.02	0.12	0.33	1.80	3.08	3.2	4.25
0.22	0.13	0.006	0.02	0	0.33	1.80	8.13	3.2	4.18
0.28	0.14	0.006	0.02	0.12	0.32	1.54	7.31	3.2	3.92
0.34	0.13	0.006	0.02	0.12	0.31	1.30	6.58	3.2	3.50
0.34 <sup>b</sup>	0.13	0.006	0.02	0.12	0.28	1.39	6.25	2.0	3.34
0.34 <sup>b</sup>	0.13	0.006	0.02	0	0.28	1.40	6.32	2.0	3.36

<sup>a</sup>All flow rates in g/s.<sup>b</sup>wedge at 0.64 cm position.

TABLE 2. N<sub>2</sub>(A) to NO Transfer Data .

Wedge	Primary		Trip	Secondary		(Molecules/cm <sup>3</sup> )				
	25% F <sub>2</sub> in HE <sup>a</sup>	Combustor D <sub>2</sub> <sup>a</sup>		He <sup>a</sup>	N <sub>2</sub> F <sub>4</sub> <sup>a</sup>	N <sub>2</sub> (B) x10 <sup>10</sup>	NO(A) x10 <sup>10</sup>	NF(a <sup>1</sup> Δ) x10 <sup>15</sup>	NF(b <sup>1</sup> Δ) x10 <sup>12</sup>	
0.183	0.12	0.007	0.02	0.03	0.21	2.23	9.23	5.2	2.71	
0	0.12	0.007	0.02	0.03	0.21	6.00	0	5.2	2.85	
0.238	0.13	0.007	0.02	0.03	0.24	1.30	2.31	4.0	1.12	
0	0.13	0.007	0.02	0.03	0.24	4.31	0	4.0	2.15	
0.33	0.12	0.007	0.02	0.03	0.23	0.54	1.22	3.9	0.73	
0	0.12	0.007	0.02	0.03	0.23	4.11	0	3.9	1.96	

<sup>a</sup>All flow rates in g/s.

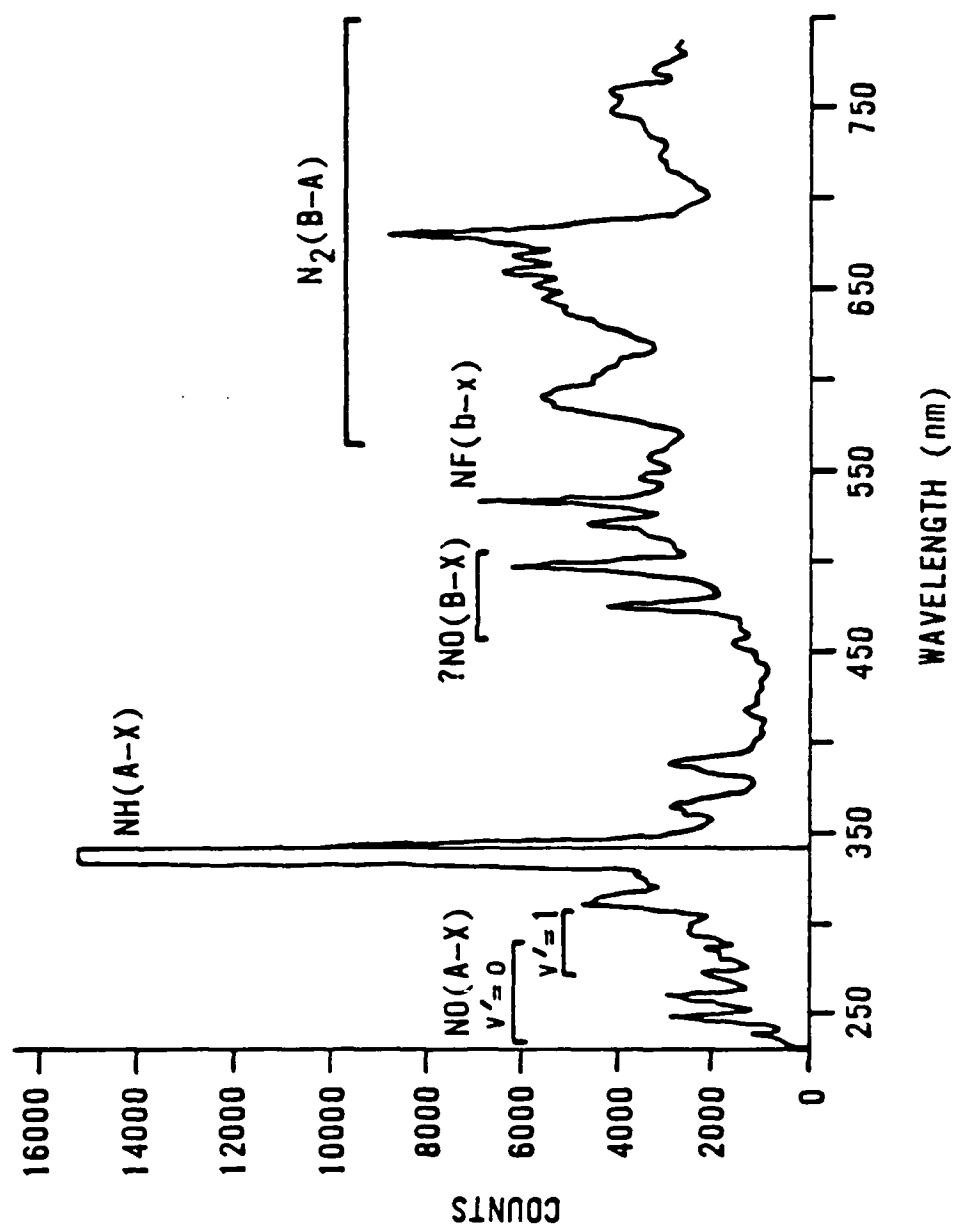


Figure 11. OMA III scan with NO flow on.

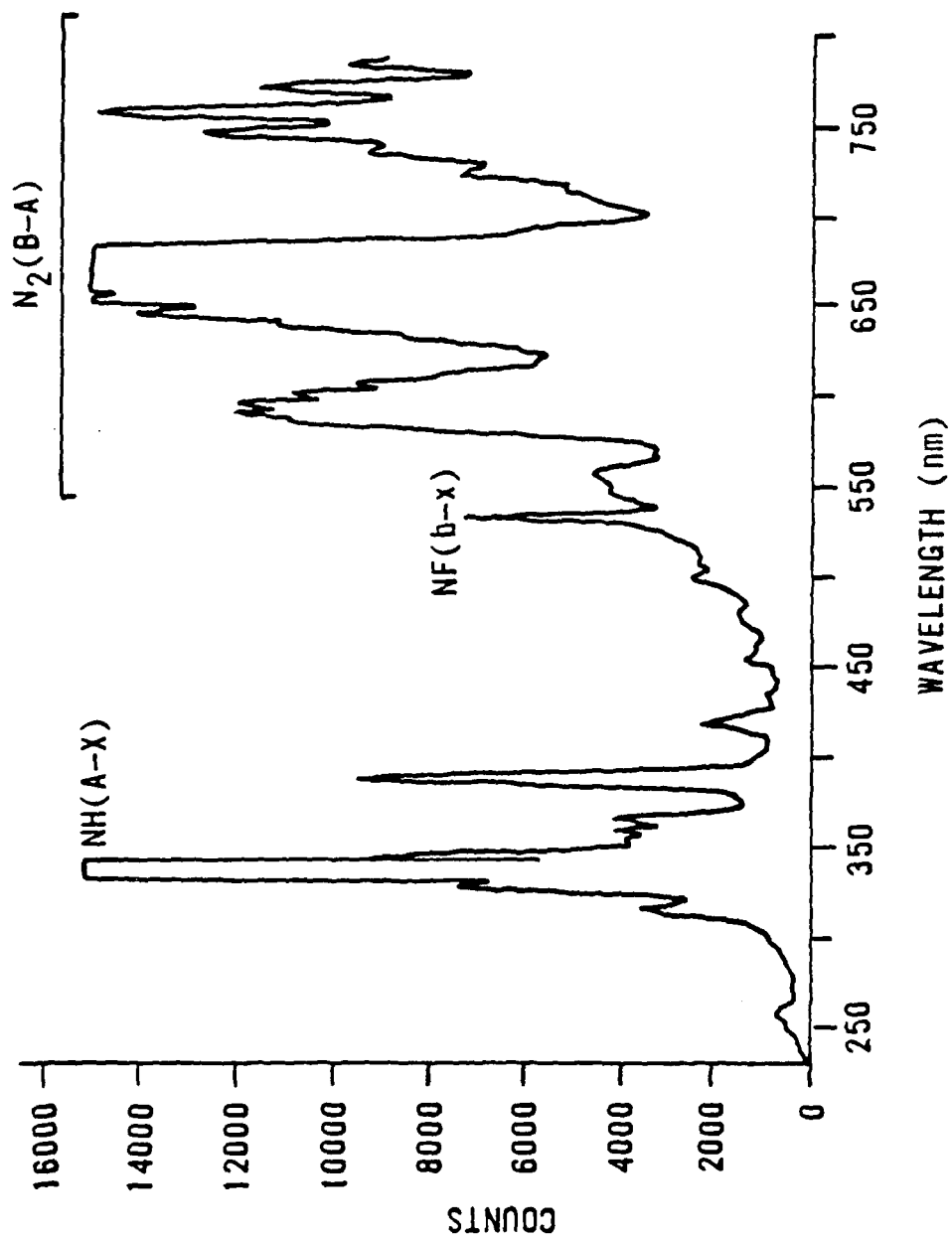


Figure 12. OMA III scan without NO flow.

## CONCLUSIONS

Efficient  $N_2(A^3\Sigma)$  to  $NO(X^2\Pi, v'' = 0)$  transfer has been demonstrated using a purely chemical production method for  $N_2(A^3\Sigma)$ . The  $N_2(A)$  production system is complex and is not completely understood. Improvements in nozzle design could increase the  $N_2(A)$  production which would allow for greater  $NO(A)$  densities. Improvements in the  $N_2(A)$  production have been discussed in Reference 21. The maximum production level of  $NO(A)$  in these experiments was about  $10^{11}$  molecules/cm<sup>3</sup>. Levels on the order of  $10^{14}$  are required to demonstrate gain over the short path length provided by this nozzle and system. Therefore, for nozzle development not only increased  $N_2(A)$  production should be examined, but also gain path length. The  $N_2(A)$  to  $NO(X)$  energy transfer system is promising for development into a chemical laser system.

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APPENDIX  
N<sub>2</sub>(A) to NO(X)  
KINETICS

TABLE A-1. Kinetic Rates.

1.	$N_2(A) + NO(X) \rightarrow N_2(X) + NO(A)$	$k_1 = 1.5 \times 10^{-10}$	(Ref 1)
2.	$NO(A) + NO(X) \rightarrow NO(X) + NO(X)$	$k_2 = 1.7 \times 10^{-10}$	(Ref 2)
3.	$NO(A) + N_2(X) \rightarrow NO(X) + N_2(X)$	$k_3 = 1.1 \times 10^{13}$	(Ref 2)
4.	$NO(A) + N_2(B) \rightarrow NO(X) + N_2(A)$	$k_4 = 2.4 \times 10^{-10}$	(Ref 3)
4a.	$\rightarrow NO(X) + N_2(X)$		
5.	$N(^2D) + NO(X) \rightarrow N_2(X) + O(^3P)$	$k_5 = 7.0 \times 10^{-11}$	(Ref 4)
5a.	$\rightarrow N_2(X) + O(^1D)$		
5b.	$\rightarrow N_2(X) + O(^1S)$		
6.	$N(^4S) + NO(X) \rightarrow N_2(X) + O(^3P)$	$k_6 = 3.6 \times 10^{-11}$	(Ref 5)
6a.	$\rightarrow N_2(X) + O(^1D)$		
6b.	$\rightarrow N_2(X) + O(^1S)$		
7.	$N_2(A) + O(^3P) \rightarrow N_2(X) + O(^3P)$	$k_7 = 2.8 \times 10^{-11}$	(Ref 6)
7a.	$\rightarrow N_2(X) + O(^1D)$		
7b.	$\rightarrow N_2(X) + O(^1S)$	$k_{7b}/k_7 = .75$	(Ref 7)
7c.	$\rightarrow NO(X) + N(^4S)$		
7d.	$\rightarrow NO(X) + N(^2D)$		
8.	$O(^1D) + N_2(X) \rightarrow O(^3P) + N_2(X)$	$k_8 = 6.9 \times 10^{-11}$	(Ref 8)
9.	$O(^1S) + O(^3P) \rightarrow O(^1D) + O(^3P)$	$k_9 = 1.8 \times 10^{-11}$	(Ref 9)
9a.	$\rightarrow O(^3P) + O(^3P)$		
10.	$O(^1D) + NO(X) \rightarrow O(^3P) + NO$	$k_{10} = 8.5 \times 10^{-11}$	(Ref 10)
10a.	$\rightarrow N(^4S) + O_2(X)$		
11.	$O(^1S) + NO(X) \rightarrow O(^1D) + NO(X)$	$k_{11} = 4 \times 10^{-10}$	(Ref 11)
11a.	$\rightarrow O(^3P) + NO(X)$		
11b.	$\rightarrow N(^2D) + O_2(X)$		
11c.	$\rightarrow N(^4S) + O_2(a)$		
11d.	$\rightarrow N(^4S) + O_2(b)$		

All rates are in  $cm^3/molecule - s$ .

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