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Weapons and Materials Research Directorate, ARL

Abstract

An experimental and chemical modeling study of neat and NH_3-doped $H_2/N_2O/Ar$ flames is conducted in order to understand the fundamental mechanism for NO formation and destruction and to predict the efficacy of NH_3 on the rate of conversion of NO to N_2. Species concentration and temperature profiles are measured with molecular-beam-mass spectrometer and thin-wire thermocouple, respectively. Species profiled include H_2,N_2O, NH_3, N_2, NO, and Ar. The experimental mole fractions are compared to both equilibrium and one-dimensional premixed laminar flame code (PREMIX) calculations. The PREMIX code employs a chemical mechanism consisting fo 87 reactions and 20 species with rate constants obtained from a critical literature review. Equilibrium claculations are in very good agreement with both experimental and PREMIX calculations for N_2O, N_2, and H_2O in the postflame region of both neat and doped flaes, but underpredict the $H_2$and NO mole fractions. The PREMIX profiles of the majority species agree very well with the experiment for the neat flame and reasonably well for the doped flame. A 55% reduction in the NO mole fraction for 4% dopant is predicted in the post-flame region, in good agreement with that observed experimentally. The flame calculations overpredict, however, the NH_3 mole fractions in the post-flame region, suggesting that refinements in the model are necessary. Rate and sensitivity analyses reveal that the decrease in NO mole fraction results from less NO formation by the reactions involving N_2O+H and more of its consumption to N_2 by reactions involving NO+NH_2.

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1. INTRODUCTION

An understanding of the way additives alter the chemical pathways in combustion systems is a prerequisite for controlling and enhancing system performance. In many systems, NO is a product of incomplete combustion whose formation prevents full energy release of the system. It is also known to cause the formation of a dark zone in the burning of solid propellants, which is undesirable [1], and, as an environmental pollutant, it poses problems in the incineration of aged propellants. Thus, it is of interest to investigate the incorporation of various additives in nitrogenous combustion systems for NO reduction. The H_2/N_2O chemical system was chosen because it is fairly simple and has important implications in understanding NO_x pollutant formation and nitramine propellant combustion and decomposition. The elementary reactions of the system are a subset of larger mechanisms needed to understand the nitrogen chemistry of more complex combustion systems.

This report describes combined experimental and modeling flame-structure studies of neat and NH_3-doped $H_2/N_2O/Ar$ flames in order to explore the role of the additive in converting NO to final products. NH_3 was selected as the additive because of its proven use in the thermal $deNO_x$ process to remove NO from effluent streams of industrial furnaces [2]. The experimental mole fractions are compared with both equilibrium and one-dimensional premixed laminar flame code (PREMIX) calculations. Rate and sensitivity analyses of the PREMIX calculations reveal the mechanisms responsible for NO formation and consumption.

2. EXPERIMENTAL

The details of the experimental apparatus used in this study are discussed elsewhere [3, 4]. Briefly, a 30-torr $H_2/N_2O/Ar$ flame, generated by flowing H_2, N_2O, and Ar at 1.78, 1.64, and 1.23 slpm, respectively, was stabilized on a 6-cm diameter flat-flame burner and doped with up to 7% NH_3. Profiles of H_2, N_2O, NH_3, H_2O, N_2, NO, and Ar were obtained by molecular-beam mass spectrometry (MBMS), and the species concentrations were determined by direct calibration at ambient temperature by a procedure previously reported [4]. For H_2O, the signals measured in the flame were quantified by equating the ratio of N/O in the premixed gases to the ratio of N/O in the burnt gases and by assuming that the mole fractions of the radical species are comparatively small [5]. The errors associated with the absolute species concentrations were ±10%. The OH, NH_2, NH, and O radicals were not measured by MBMS because of the same mass-to-charge ratio and fragmentation-ionization interferences resulting from the addition of NH_3. These species are, however, presently being measured by laser-induced fluorescence (LIF), and the results will be presented elsewhere.

Flame temperatures were measured with a Pt/Pt-Rh(10%) fine-wire thermocouple coated with a beryllium oxide (15%)/yttrium oxide mixture [6] and corrected for temperature-radiation losses using OH LIF [7]. The overall uncertainty of temperature measurements is ±100 K in the region of the peak temperature.

The temperature data for both neat and NH_3-doped $H_2/N_2O/Ar$ flames, measured near the quartz sampler and fit to a sigmoid-type function, are shown in Figure 1. The peak in the temperature profiles near the burner surface is an artifact produced by the quartz sampler since it is not present when the sampler is removed. (Temperature profiles without sampler are not shown.) Doping the neat flame with 4% of NH_3 results in approximately a 2-mm displacement of the temperature profile away from the burner surface, and approximately a 44-K drop in the peak temperature. The small drop in peak temperature is consistent with NASA-Lewis equilibrium calculations [8], which yield a difference in the adiabatic flame temperatures of only 9 K. In the post-flame region, the measured flame temperatures are nearly identical, approximately 1,822 K at 13.75 mm.

3. MODELING/CHEMICAL MECHANISM

The computations utilized the Sandia Laboratories PREMIX code (ver.2.5b) [9], which employs the CHEMKIN-II (ver. 3.0) library [10]. The calculations were performed with the fitted- temperature profiles used as input to PREMIX, the inclusion of both thermal diffusion and multicomponent transport package option, and with normal boundary conditions (i.e., no

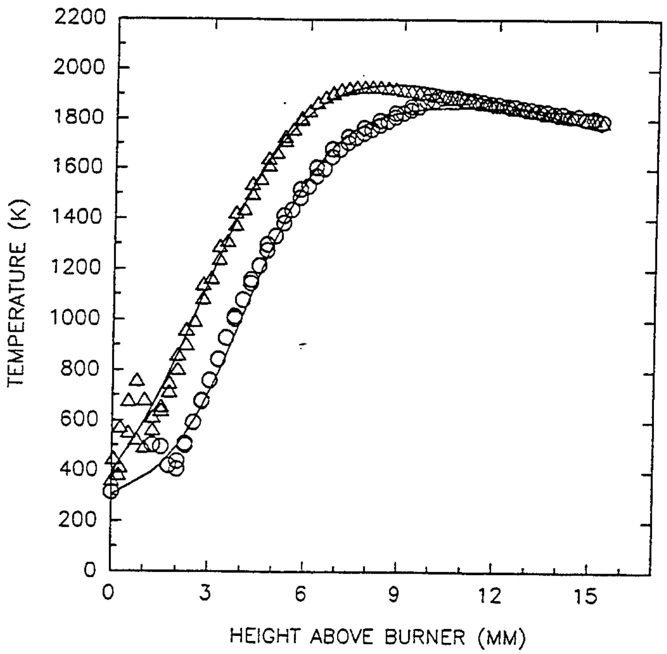

Figure 1. Temperature profiles of a neat (Δ) and 4% NH_3-doped (o), 30-torr $H_2/N_2O/Ar$ flame. The solid line is a best fit of the data using a sigmoid-type function.

recombination of H atoms to H_2 via burner surface reaction was included). Prior work under conditions similar to those used here indicates that inclusion of this recombination effect only results in a minor change to the H and H_2 profiles very close to the burner surface [3]. Gridding parameters and calculational domain were checked to ensure that numerical errors due to resolution and hot boundary conditions were negligible. Transport and thermochemical databases [11, 12] developed at Sandia were used for these calculations with the exception of a few of the species' heats of formation. Values of $(\Delta H^o_f)_{298}$ for HNO, NH_2, and NH were updated to 25.4, 45.2, and 85.3 kcal/mole, respectively [13, 14]. The program THERM [15] was used to refit these species' thermal data to the form used in CHEMKIN codes. Rate and sensitivity analyses were performed using an interactive postprocessing code written in-house.

The chemical mechanism and rate constants used for the flame calculations are presented in Table 1. The bulk of the reactions was obtained from the benchmark review of nitrogen chemistry in combustion by Miller and Bowman [16], which were updated for the H_2/N_2O system [3, 4]. More recent updates include both rate constant [25] and efficiency factor

Table 1. Reaction Mechanism Employed for Modeling Both Neat and NH_3-Doped H_2/N_2O Flames (The Rate Coefficients Are in the Form $k=AT^Be^{(-E/RT)}$, Where *A* and E Have Units of cm-mol-s-K and cal/mol, Respectively)

	REACTIONS	A	B	E	REF.
1	H2+O2=OH+OH	1.70E+13	0.00	47780.0	[16]
2	H2+OH=H2O+H	2.16E+08	1.51	3430.0	[17]
3	H2+O=H+OH	5.06E+04	2.67	6290.0	[18]
4	H+H+H2=H2+H2	1.00E+17	-0.60	0.0	[19]
5	H+H+H2O=H2+H2O	1.00E+19	-1.00	0.0	[19]
6	H+H+H=H2+H	3.20E+15	0.00	0.0	[19]
7	H+H+AR=H2+AR	7.00E+17	-1.00	0.0	[19]
8	H+H+N2=H2+N2	5.40E+18	-1.30	0.0	[19]
9	H+H+M=H2+M	1.00E+18	-1.00	0.0[a]	[16]
10	H+O2=OH+O	3.52E+16	-0.70	17070.0	[20]
11	O2+H+M=HO2+M	3.61E+17	-0.72	0.0[a]	[16]
12	O+O+M=O2+M	1.89E+13	0.00	-1788.0	[16]
13	HO2+OH=H2O+O2	7.50E+12	0.00	0.0	[16]
14	HO2+H=OH+OH	1.69E+14	0.00	874.0	[21]
15	HO2+H=H2+O2	6.63E+13	0.00	2126.0	[21]
16	HO2+O=OH+O2	1.40E+13	0.00	1073.0	[16]
17	OH+OH=H2O+O	6.00E+08	1.30	0.0	[16]
18	H+OH+M=H2O+M	2.20E+22	-2.00	0.0[a]	[22]
19	H+O+M=OH+M	6.20E+16	-0.60	0.0[a]	[16]
20	N2O+H=OH+N2	2.53E+10	0.00	4550.0[b]	[23]
		2.23E+14	0.00	16750.0[b]	[23]
21	NNH+O=N2O+H	1.40E+14	-0.40	477.0	[24]
22	N2O+M=N2+O+M	5.97E+14	0.00	56640.0[a]	[25]
23	N2O+O=N2+O2	1.40E+12	0.00	10800.0	[26]
24	N2O+O=NO+NO	6.90E+13	0.00	26600.0	[27]
25	NH+NO=N2O+H	2.94E+14	-0.40	0.0[b]	[28]
		-2.16E+13	-0.23	0.0[b]	[28]
26	NH+NO=N2+OH	2.16E+13	-0.23	0.0	[28]
27	NH+O2=NO+OH	7.60E+10	0.00	1530.0	[29]
28	NH+O2=HNO+O	3.89E+13	0.00	17885.0	[29]
29	NH+OH=HNO+H	2.00E+13	0.00	0.0	[16]
30	NH+OH=N+H2O	5.00E+11	0.50	2000.0	[16]
31	NH+N=N2+H	3.00E+13	0.00	0.0	[16]
32	N+H2=NH+H	1.60E+14	0.00	25140.0	[30]
33	NH+O=NO+H	5.50E+13	0.00	0.0	[29]
34	NH+O=N+OH	3.72E+13	0.00	0.0	[29]
35	NH+NH=N2+2H	5.10E+13	0.00	0.0	[31]
36	NH+M=N+H+M	2.65E+14	0.00	75514.0	[31]
37	NH2+O=HNO+H	4.60E+13	0.00	0.0	[32]
38	NH2+O=NH+OH	7.00E+12	0.00	0.0	[32]
39	NH2+OH=NH+H2O	4.00E+06	2.00	1000.0	[16]
40	NH2+H=NH+H2	4.00E+13	0.00	3650.0	[33]
41	NH2+NO=NNH+OH	6.40E+15	-1.25	0.0	[16]
42	NH2+NO=N2+H2O	6.20E+15	-1.25	0.0	[16]
43	NH2+NH=N2H2+H	1.50E+15	-0.50	0.0	[33]
44	NH2+N=N2+H+H	7.20E+13	0.00	0.0	[16]
45	NH2+O2=HNO+OH	4.50E+12	0.00	25000.0	[16]
46	NH2+NH2=N2H2+H2	5.00E+11	0.00	0.0	[16]
47	NH2+NH2=NH+NH3	5.00E+13	0.00	10000.0	[33]
48	NH3+OH=NH2+H2O	2.04E+06	2.04	566.0	[16]
49	NH3+H=NH2+H2	5.42E+05	2.40	9917.0	[34]
50	NH3+O=NH2+OH	9.40E+06	1.94	6460.0	[35]
51	NH3+M=NH2+H+M	2.20E+16	0.00	93470.0	[33]
52	NNH+NO=N2+HNO	5.00E+13	0.00	0.0	[16]
53	NNH+H=N2+H2	1.00E+14	0.00	0.0	[16]
54	NNH+OH=N2+H2O	5.00E+13	0.00	0.0	[16]
55	NNH+NH2=N2+NH3	5.00E+13	0.00	0.0	[16]
56	NNH+NH=N2+NH2	5.00E+13	0.00	0.0	[16]
57	NNH=N2+H	3.00E+08	0.00	0.0	[24]
58	NNH+M=N2+H+M	1.00E13	0.50	3060.0	[24]
59	NNH+O=N2+OH	1.70E16	-1.23	497.0	[24]
60	NNH+O=NO+NH	3.30E14	-0.23	-1013.0	[24]
61	HNO+OH=NO+H2O	1.30E+07	1.88	-960.0	[36]
62	HNO+NH2=NH3+NO	2.00E+13	0.00	1000.0	[16]
63	HNO+HNO=N2O+H2O	3.63E-03	3.98	1190.0	[37]
64	HNO+NO=N2O+OH	8.51E+12	0.00	29590.0	[38]
65	HNO+0=OH+NO	4.50E11	0.72	656.0	[32]
66	N2H2+M=NNH+H+M	5.00E+16	0.00	50000.0[a]	[16]
67	N2H2+H=NNH+H2	5.00E+13	0.00	1000.0	[16]
68	N2H2+O=NH2+NO	1.00E+13	0.00	0.0	[16]
69	N2H2+O=NNH+OH	2.00E+13	0.00	1000.0	[16]
70	N2H2+OH=NNH+H2O	1.00E+13	0.00	1000.0	[16]
71	N2H2+NH=NNH+NH2	1.00E+13	0.00	1000.0	[16]
72	N2H2+NH2=NH3+NNH	1.00E+13	0.00	1000.0	[16]
73	HNO+H=H2+NO	4.46E+11	0.72	655.0	[39]
74	NO+N=N2+O	3.27E+12	0.30	0.0	[16]
75	NO+M=N+O+M	1.40E+15	0.00	148430.0	[40]
76	NO+H+M=HNO+M	9.00E+19	-1.30	735.0	[40]
77	NO+H=N+OH	1.70E+14	0.00	48801.0	[27]
78	NO+O=N+O2	3.80E+09	1.00	41370.0	[40]
79	NH2+NH2=N2H3+H	1.79E+13	-0.35	11320.0	[32]
80	NH2+NH2+M=N2H4+M	2.98E+47	-9.44	9680.0	[32]
81	N2H4+H=N2H3+H2	7.05E+12	0.00	2500.0	[41]
82	N2H4+OH=N2H3+H2O	3.00E+10	0.68	1290.0	[42]
83	N2H4+O=N2H3+OH	2.00E+13	0.00	1000.0	[42]
84	N2H3+M=N2H2+H+M	1.20E+13	0.00	58000.0	[42]
85	N2H3+H=N2H2+H2	1.00E+12	0.50	2000.0	[42]
86	N2H3+OH=N2H2+H2O	3.00E+10	0.68	1290.0	[42]
87	N2H3+O=N2H2+OH	2.00E+13	0.00	1000.0	[42]

[a] Third body efficiencies.
R9. H_2/0.0/H_2O/0.0/H/0.0/Ar/0.0/N_2/0.0
R11. N_2/1.3/H_2O/18.6/H_2/2.9
R18. Ar/0.38/H_2O/6.3
R19. H_2O/5.0
R22. N_2O/5.0/H_2O/7.5/N_2/1.0/O_2/.82/Ar/0.67
R66. H_2O/15.0/O_2/2.0/N_2/2.0/H_2/2.0

[b] For the indicated reactions the rate coefficient is computed as the sum of the two exponential expressions given for each.

($M=H_2O$) [43] for the sensitive reaction $N_2O+M=N_2+O+M$ (R22). Recent results of Hanson and coworkers for many important reactions involving NH_x species that were previously poorly quantified have also been incorporated [29–31, 33]. Finally, the reaction $OH+N_2O=HO_2+N_2$ was not included in the mechanism since recent experimental [3, 43, 44] and theoretical [45] studies indicate it is too slow to be of importance.

4. RESULTS/DISCUSSION

The experimental and predicted mole fractions of H_2, N_2, H_2O, and N_2O for the neat $H_2/N_2O/Ar$ flame are presented in Figure 2. Also presented are species profiles generated by increasing the temperature profile by a factor of 1.05, the experimental uncertainty. The computed profiles accurately model the experimental results throughout the entire flame for the neat flame. Quantitatively, the calculated mole fractions agree well with those measured experimentally, particularly in the post-flame region. NASA-Lewis calculations performed using a temperature of 1,822 K yield equilibrium mole-fraction values of 0.351 for N_2 and H_2O and 0.0 for N_2O, which are also in good agreement with the experimental mole fractions at 13.75 mm. The calculated H_2 equilibrium value of 0.0313 is, however, a factor of 2 smaller than the experimental value, indicating incomplete reaction.

Figure 3 shows the experimental and calculated mole-fraction profiles of the major species for the NH_3-doped flame. Species profiles generated by increasing the experimental temperature profile by the experimental uncertainty are also presented. Both the experimental and modeled profiles are shifted away from the burner compared to their positions in the neat flame. The result follows the trend in temperature profiles (Figure 1). For the doped flame, the modeled profiles are shifted approximately 2 mm further away from the burner surface compared to those measured, indicating that the overall chemical reaction rates in the model are too slow. The experimental and calculated mole-fraction values for H_2, H_2O, and N_2 agree reasonably well, while the N_2O profile is overpredicted by the model. As in the neat flame, equilibrium mole-

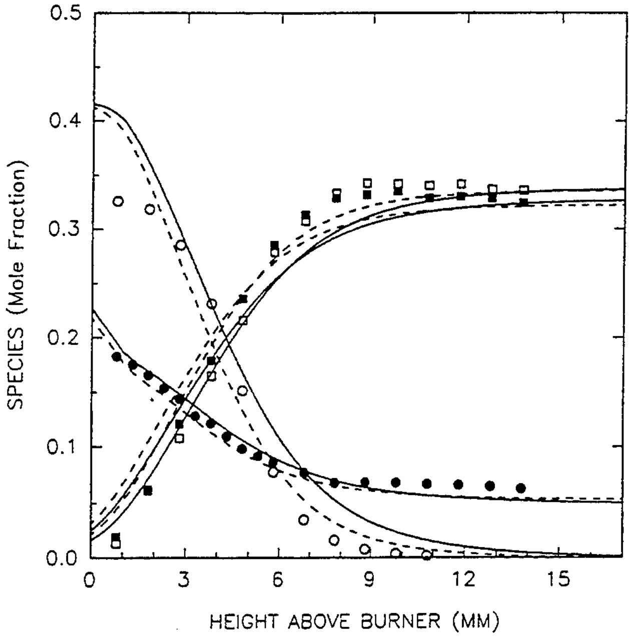

Figure 2. Calculated (–) and experimental H_2 (●), N_2O (o), H_2O (■), and N_2 (□) species profiles for a 30-torr $H_2/N_2O/Ar$ flame. The dashed curves are calculated profiles generated by increasing the temperature profile by a factor of 1.05, the experimental uncertainty.

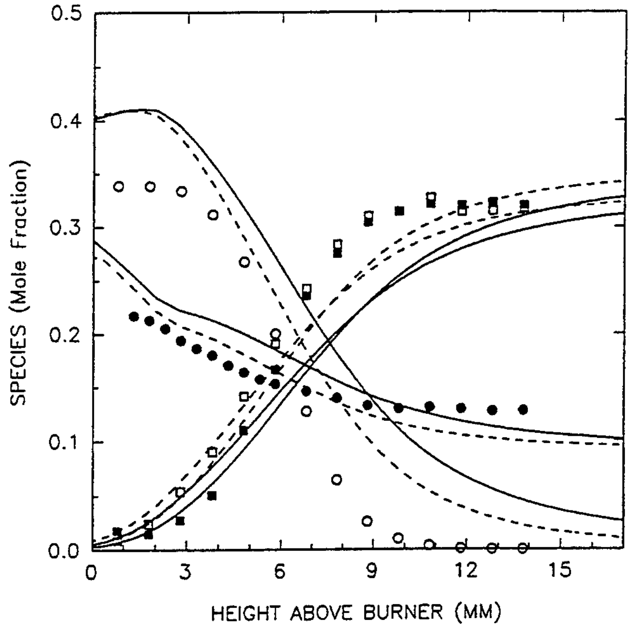

Figure 3. Calculated (–) and experimental H_2 (●), N_2O (o), H_2O (□), and N_2 (■) species profiles for a 30-torr $H_2/N_2O/Ar$ flame doped with 4% NH_3. The dashed curves are calculated profiles generated by increasing the temperature profile by a factor of 1.05, the experimental uncertainty.

fraction values for N_2, H_2O, and N_2O are in good agreement with the experimental values at 13.75 mm. The equilibrium H_2 mole fraction is, however, a factor of 1.4 lower than the experimental value.

The calculated and experimental NO mole-fraction profiles for both the neat and NH_3-doped flames are shown in Figure 4. Quantitatively, excellent agreement between model and experiment is obtained in the post-flame region. The addition of 4% NH_3 reduces the NO concentration by approximately 45%, an effect predicted well by the model. Also, a leveling-off of the reduction efficiency was observed experimentally for NH_3 concentrations greater than approximately 6%. A 5% increase in the temperature profile inputted to the model resulted in approximately a 15 and 20% increase in the calculated NO mole fraction for neat and doped flames, respectively. As shown in Figure 4, the model and experimental NO profile shapes are similar. However, the modeled NO profile for the doped flame peaks at 9 mm and gradually decreases thereafter, in contrast to the experimental profile that plateaus at 9–10 mm. This discrepancy is related to an overprediction of NH_3 in the post-flame region by the model, which will be discussed in the following paragraph. Results of equilibrium calculations for NO at 13.75 mm are three orders of magnitude lower than both PREMIX and experimental values, indicating that NO in the post-flame region is well above its equilibrium concentration.

Also presented in Figure 4 are the experimental and calculated mole-fraction profiles of NH_3 for the doped flame. (The NH_3 mole fraction in the neat flame is negligibly small.) The modeled profile represented by the solid line strongly overpredicts the NH_3 mole fraction in the burnt gases. Equilibrium calculations predict the NH_3 mole fraction to be negligible at 13.75 mm, as observed experimentally. Also shown in Figure 4 are modeled profiles of NH_3 for (1) a 5% increase in the entire temperature profile, and (2) an assumed area expansion coefficient of 0.25 cm^{-1} as well as a 5% increase in the temperature profile. The profile generated with the latter conditions is in close agreement with the experimental profile in the lower part of the flame, but still overpredicts the experimental profile at distances greater than 6 mm. Although visual inspection indicated the expansion coefficient was no larger than 0.25 cm^{-1}, values of 0.50 and 1.00 cm^{-1} were also tried in the model. In all cases, the predicted NH_3 concentration in

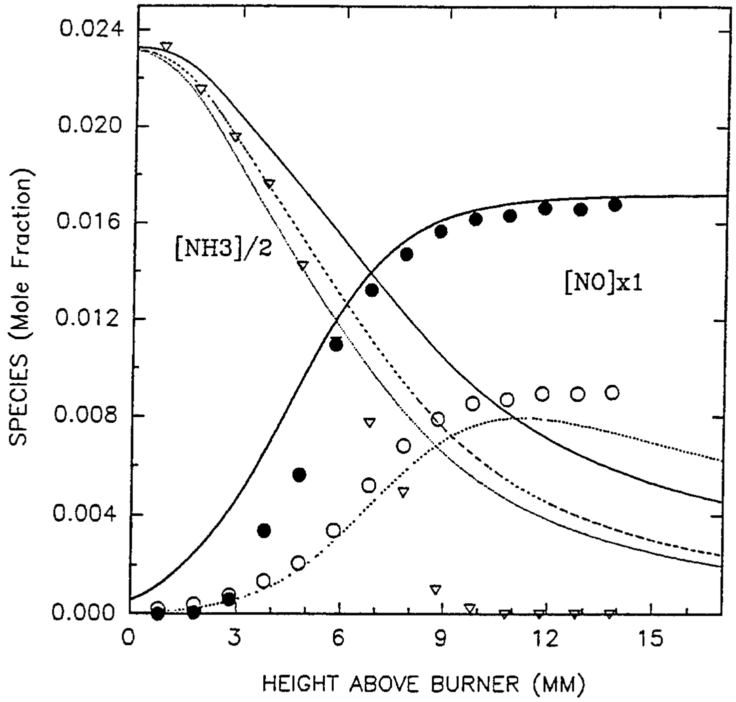

Figure 4. Calculated (–) and experimental NO profiles for both neat (●) and 4% NH_3-doped (o) $H_2/N_2O/Ar$ flames, and experimental (▽) and calculated (–) NH_3 profiles for doped flame. The experimental NH_3 profile is normalized to that which is calculated. Also shown are NH_3 PREMIX profiles generated by (a) increasing the measured temperature profile by 5% (---) and (b) increasing the temperature profile by 5% and using $A = 1 + 0.25x$ for the area expansion term, where x is the distance from the burner surface (...). The pressure was maintained at 30 torr.

the burnt gases was far above that permitted by experimental limits. A factor of 10 reduction in the NH_3 mole fraction was obtained, however, by altering simultaneously the rate coefficients of the ten most sensitive reactions for NH_3 (see Table 2) by a factor of two in the direction that the analysis indicated would reduce the predicted NH_3 burnt gas mole fraction. However, making these changes simultaneously is rather drastic and quite unlikely to be correct since the resulting rate constants are close to or exceed the error limits for most of the reactions. The result is still far higher than the experimental limits will allow, particularly at 12–14 mm. Reaction flux analysis in the burnt gases (not shown) demonstrates conclusively that this overpredicted NH_3 mole fraction is the cause of the predicted NO profile decay in the burnt gases, which, as mentioned previously, disagrees with the experiment. In the model, NH_3 in this region reacts to

Table 2. Sensitivity Coefficients for NH_3 in NH_3-Doped Flame at 3.75 mm Above the Burner Surface

REACTION		SENSITIVITY COEFFICIENTS (Rel.)	
		Sign	Doped[a]
22	N2O+M=N2+O+M	(-)	100.0
20	N2O+H=OH+N2	(+)	62.6
43	NH2+NH=N2H2+H	(-)	36.2
48	NH3+OH=NH2+H2O	(-)	32.7
2	H2+OH=H2O+H	(+)	22.1
41	NH2+NO=NNH+OH	(-)	16.0
25	NH+NO=N2O+H	(-)	14.7
60	NNH+O=NO+NH	(-)	12.9
32	N+H2=NH+H	(+)	11.6
29	NH+OH=HNO+H	(+)	10.7
40	NH2+H=NH+H2	(-)	8.8
66	N2H2+M=NNH+H+M	(-)	8.7
49	NH3+H=NH2+H2	(+)	7.9
44	NH2+N=N2+H+H	(-)	7.3
67	N2H2+H=NNH+H2	(+)	6.7
77	NO+H=N+OH	(+)	6.4
37	NH2+O=HNO+H	(+)	4.2
26	NH+NO=N2+OH	(+)	4.1
76	NO+H+M=HNO+M	(-)	4.0

[a] Logarithmically normalized [3] to 100 for the sensitivity coefficient of reaction 22 (−0.6993).

produce NH_x molecules (primarily NH_2) that continue to reduce the NO resulting in the decaying trend seen in Figure 4.

It should be noted that the choice of branching ratio for the NH_2+NO reaction (R41 and 42), a well-known subject of controversy, has little effect on the computed profiles. Tests have conclusively proven that the computed profiles are sensitive only to the total rate coefficient, not to the ratio. The tests were performed by adding the two rate coefficient expressions and then using the resulting rate coefficient for one of the two channels while removing the other. Under the current high-temperature conditions, N_2O+M (R22) is the primary radical source and NH_2+NO provides comparatively few radicals. In contrast, under the lower temperature thermal

$deNO_x$ conditions, the chain-branching reaction R41 must be included to provide a radical source to sustain the overall reaction [16].

Figure 5 is a reaction pathway diagram depicting the nitrogen chemistry occurring in the neat $H_2/N_2O/Ar$ flame. The numbers in parentheses are the relative integrated rates (0–13.75 mm) of the various reactions normalized to 100 (4.428×10^{-6} mole/cm^2 s) for the reaction $NH_3+OH=NH_2+H_2O$ in the doped flame. As seen from Figure 5, the reaction $N_2O+H=N_2+OH$ represents the fastest step consuming N_2O to generate N_2. The reaction $N_2O+M=N_2+O+M$ is also important. NO is formed predominantly from the reaction $N_2O+H=NO+NH$. The formation of NO from NH, via the HNO intermediate, and reaction N+OH=NO+H is also significant. The partial conversion of NO to N_2, directly or indirectly via NNH, occurs predominantly by reactions involving N and NH. Not surprisingly, the species NH_3, NH_2, NNH, and N_2H_2 do not play major roles in the neat flame.

Figure 6 is a reaction pathway diagram for the NH_3-doped flame. The addition of 4% NH_3 increases the number of reactions and species having major effects on the flame. As expected, the addition of the dopant causes only a small perturbation in the reactions involving the major species as compared to those in the neat flame. However, the addition of 4% NH_3 does significantly reduce the NO concentration. This results primarily from two effects: (1) a slight decrease in NO formation from reactions $N_2O+H=NO+NH$ and N+OH=NO+H and (2) an increase in NO consumption by the NH_2 radical. NH_2 can convert NO to N_2 directly, or indirectly via the NNH intermediate. It should be specifically noted that the lowering of NO concentration in the doped flame is primarily chemical in origin, rather than due to the slight change in temperature profiles. This was proven by switching the input temperature profiles in the calculations. Switching the profiles causes only a slight change in the predicted NO profiles, particularly in the burnt gas concentrations, for either the neat or doped flame.

Table 3 compares the sensitivities of NO to various reaction rate coefficients for both neat and NH_3-doped $H_2/N_2O/Ar$ flames. The sensitivity coefficients were calculated at 13.75 mm for both flames. They are scaled to 100 for reaction R20, $N_2O+H=OH+N_2$. Table 3 reveals that for

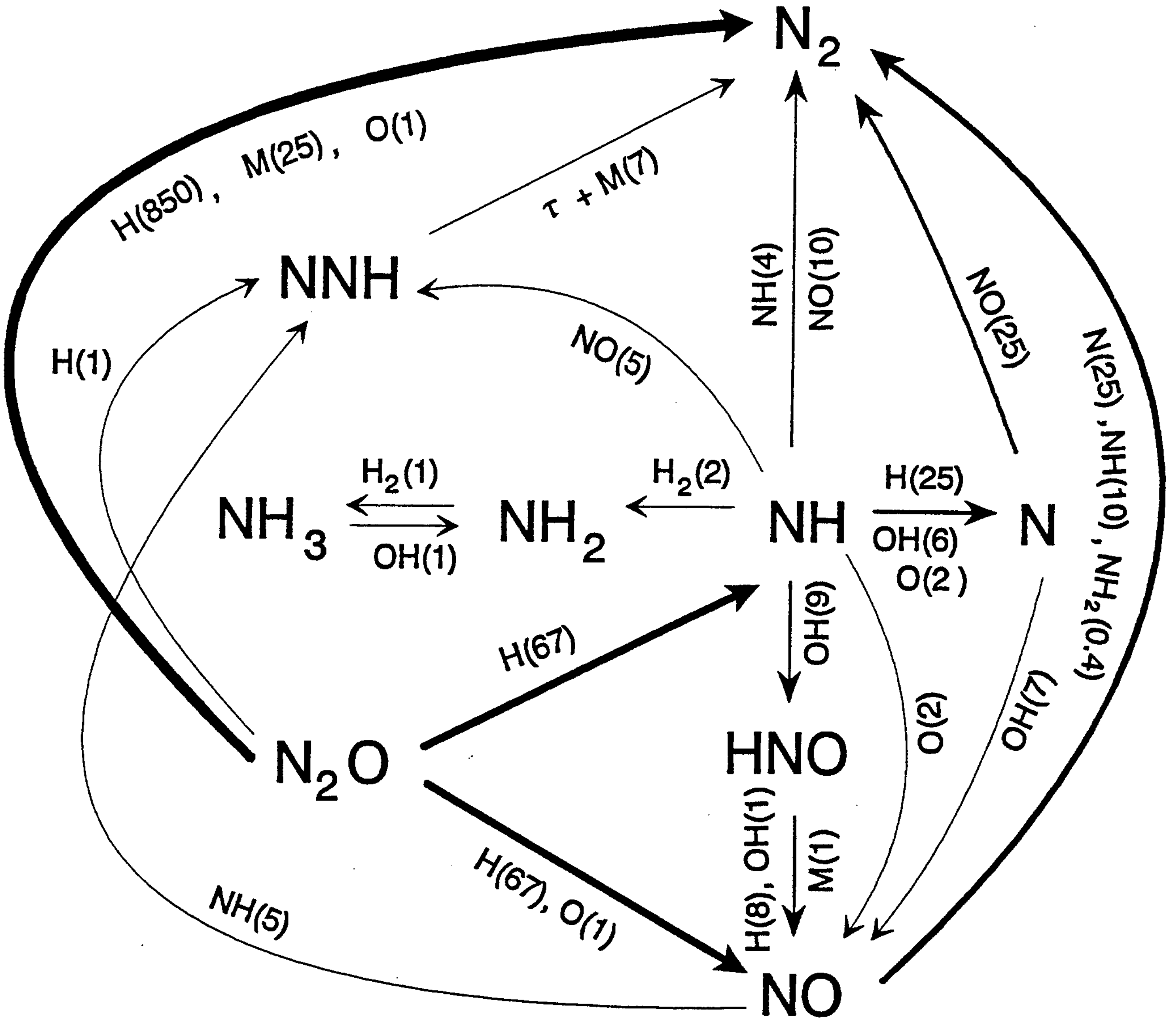

Figure 5. Pathway diagram of a 30-torr, neat $H_2/N_2O/Ar$ flame generated by integrating the net rate fluxes of individual reactions from the burner surface to 13.75 mm. For NNH destruction, τ + M indicates the total predissociation and collision assisted decomposition. (100 = 4.428×10^{-6} mole/cm^2-s).

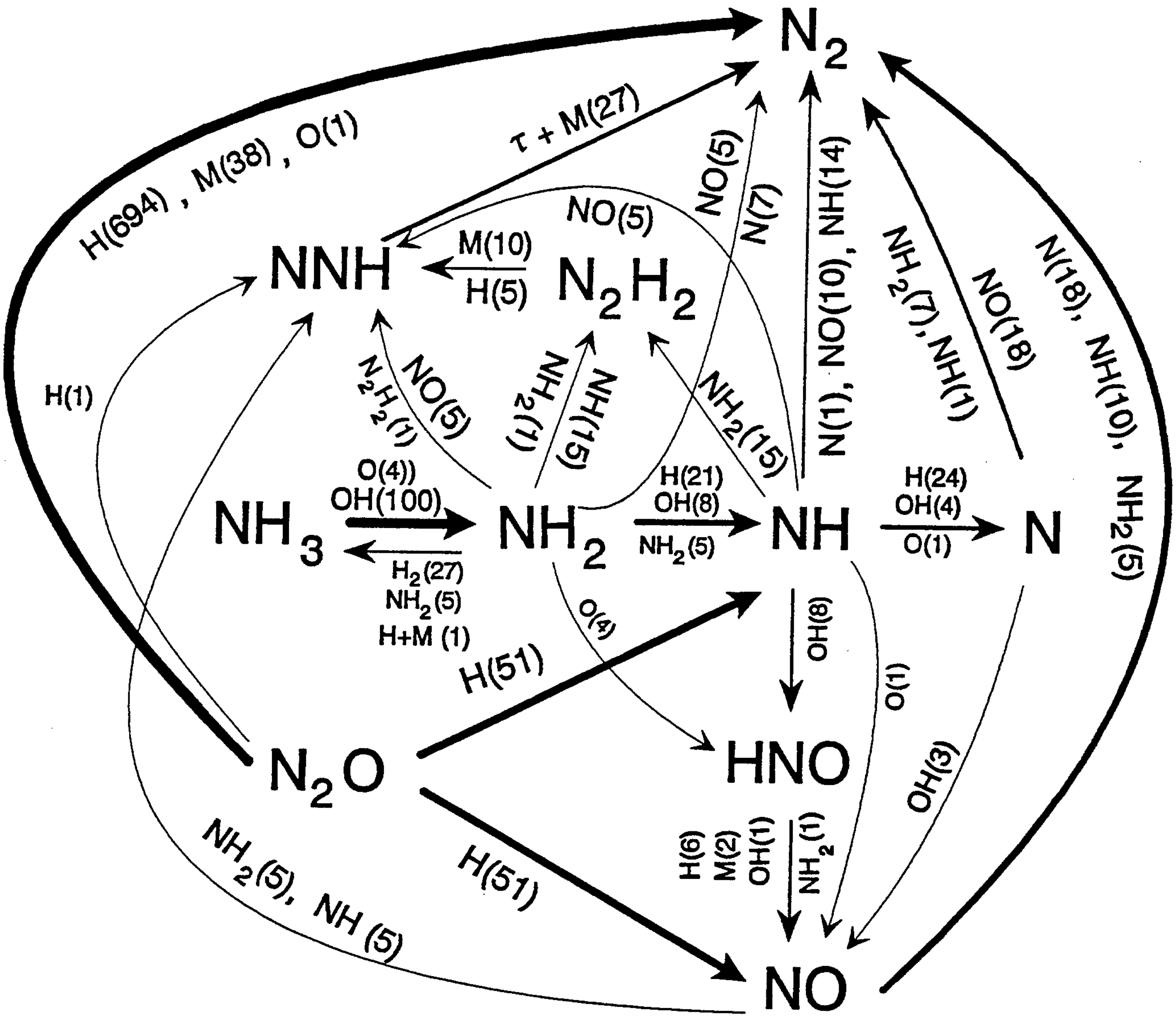

Figure 6. Pathway diagram of a 30-torr, 4% NH_3-doped $H_2/N_2O/Ar$ flame generated by integrating the net rate fluxes of individual reactions from the burner surface to 13.75 mm. For NNH destruction, τ + M indicates the total predissociation and collision assisted decomposition. ($100 = 4.428 \times 10^{-6}$ mole/cm^2-s).

Table 3. Sensitivity Coefficients for NO in a Neat and NH3-Doped Flame at 13.75 mm Above the Burner Surface

	REACTION		SENSITIVITY COEFFICIENTS (Rel.)	
		Sign	Neat[a]	Doped[b]
20	N2O+H=OH+N2	(-)	100.0	100.0
25	NH+NO=N2O+H	(+)	60.2	71.4
2	H2+OH=H2O+H	(-)	25.6	21.0
29	NH+OH=HNO+H	(+)	25.5	22.7
74	NO+N=N2+O	(-)	21.5	26.5
77	NO+H=N+OH	(+)	20.3	14.1
22	N2O+M=N2+O+M	(+)	15.0	3.4
26	NH+NO=N2+OH	(-)	10.6	19.3
33	NH+O=NO+H	(+)	6.4	4.6
35	NH+NH=N2+2H	(+)	3.7	11.5
3	H2+O=H+OH	(-)	3.4	8.8
24	N2O+O=NO+NO	(+)	2.7	1.7
32	N+H2=NH+H	(-)	2.6	4.7
60	NNH+O=NO+NH	(-)	1.7	7.9
37	NH2+O=HNO+H	(+)	1.3	10.4
43	NH2+NH=N2H2+H	(+)	<1	20.4
41	NH2+NO=NNH+OH	(-)	<1	11.3
42	NH2+NO=N2+H2O	(-)	<1	10.9
44	NH2+N=N2+H+H	(+)	<1	10.3
40	NH2+H=NH+H2		(+) <1	(-) 12.3
39	NH2+OH=NH+H2O	(-)	<1	4.3
49	NH3+H=NH2+H2	(-)	<1	3.1
17	OH+OH=H2O+O		(-) <1	(+) 2.3

[a] Logarithmically normalized [3] to 100 for the sensitivity coefficient of reaction 20 of neat $H_2/N_2O/Ar$ flame (-0.5764).

[b] Logarithmically normalized [3] to 100 for the sensitivity coefficient of reaction 20 of NH_3-doped $H_2/N_2O/Ar$ flame (-0.6165).

both the neat and doped flames the NO concentration has a strong negative sensitivity to R20 and strong positive sensitivity to the reaction N_2O+H=NH+NO, -R25. This result is clearly due to the competition between the two channels for the N_2O+H reaction, one forming NO, the other N_2. For the neat flame, NO exhibits lesser but still important sensitivities to reactions R2, R29, R74, R77, R22, and R26. For most of these reactions the trends are similar for the doped flame.

However, one notes that in comparison, the doped flame shows much stronger sensitivities than the neat flame to several additional reactions. In particular, note that most of these additional reactions involve NH_2, NH, or N. This is not surprising since the pathway diagrams indicate that these species play key roles in the consumption of NO. The signs of some of the sensitivity coefficients may be understood in terms of the competition between NH_x+NO reactions, which result in the destruction of NO, and NH_x+NH_y reactions, which ultimately result in conversion of the NH_3 to N_2 without removal of NO. For example, the two NH_2+NO reactions, R41 and R42, have negative coefficients because they ultimately convert NO to N_2. In contrast the NH_2+NH and NH+NH reactions, R43 and R35 respectively, merely convert two NH_x molecules ultimately to N_2 without destroying NO, thus, exhibiting a positive NO sensitivity coefficient. It appears that this competition for NH_x molecules is the reason for the leveling off of the effectiveness of NH_3 in reducing the NO that was observed experimentally. As the concentration of NH_3 is increased, the importance of NH_x+NH_y reactions increases.

5. CONCLUSION

A combined experimental and modeling flame structure study of neat and NH_3-doped $H_2/N_2O/Ar$ flames has been performed. Species' mole fractions and temperature profiles were recorded using MB/MS and thermocouple techniques, respectively. The modeling studies consisted of both equilibrium and PREMIX calculations. A chemical mechanism for the system was developed from a critical literature review and employed for modeling the species' profiles. The modeled profiles of the majority species agree very well with the experimental profiles for the neat flame and reasonably well for the doped flame. Quantitatively, the agreement between the modeled and experimental NO profiles is good for both the neat and doped flames. The modeled profile shows a 55% reduction in the NO mole fraction in the post-flame region when 4% NH_3 is added to the neat flame compared to a 45% reduction in the measured profile. The modeled and experimental NO profiles for the neat flame and the experimental profile in the doped flame exhibit plateaus in the post-flame region. However, the modeled NO profile in the doped flame exhibits a post-flame decay. In addition, the model overpredicts the NH_3 mole fraction in the post-flame region. Rate analysis shows, conclusively, that this overprediction is the primary cause of the predicted NO decay. Calculations indicate that these discrepancies are due to the model and not the experiment, suggesting that refinements in the chemical mechanism are necessary. Rate and sensitivity analyses reveal that the dopant slightly decreases the amount of NO formed by the reactions $N_2O+H=NO+NH$ and $N+OH=NO+H$ and increases its consumption via reaction with NH_2, which leads to conversion of NO to N_2. The efficacy of NH_3 in reducing NO is stymied at high NH_3 concentrations, an effect likely due to the increased role of $NH_x + NH_y$ reactions that convert NH_3 to N_2.

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R. C. Sausa, G. Singh, G. W. Lemire, and W. R. Anderson

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U.S. Army Research Laboratory
ATTN: AMSRL-WM-PC
Aberdeen Proving Ground, MD 21005-5066

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13. ABSTRACT ***(Maximum 200 words)***

understand the fundamental mechanism for NO formation and destruction and to predict the efficacy of NH_3 on the rate of conversion of NO to N_2. Species concentration and temperature profiles are measured with molecular-beam-mass spectrometer and thin-wire thermocouple, respectively. Species profiled include H_2,N_2O, NH_3, N_2, NO, and Ar. The experimental mole fractions are compared to both equilibrium and one-dimensional premixed laminar flame code (PREMIX) calculations. The PREMIX code employs a chemical mechanism consisting fo 87 reactions and 20 species with rate constants obtained from a critical literature review. Equilibrium claculations are in very good agreement with both experimental and PREMIX calculations for N_2O, N_2, and H_2O in the postflame region of both neat and doped flaes, but underpredict the $H_2$and NO mole fractions. The PREMIX profiles of the majority species agree very well with the experiment for the neat flame and reasonably well for the doped flame. A 55% reduction in the NO mole fraction for 4% dopant is predicted in the post-flame region, in good agreement with that observed experimentally. The flame calculations overpredict, however, the NH_3 mole fractions in the post-flame region, suggesting that refinements in the model are necessary. Rate and sensitivity analyses reveal that the decrease in NO mole fraction results from less NO formation by the reactions involving N_2O+H and more of its consumption to N_2 by reactions involving NO+NH_2.

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