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Computational Studies on One-Dimensional Laminar, Premixed Hydrogen-Nitric Oxide Flames

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Results of computational studies of adiabatic flame propagation in the hyperogen-nitric oxide system are presented here. The sensitivity of flame speed to the rate constants of various reactions is examined. Reactions of the extended Zeldovich mechanism $H + NO = N + OH$ and $N + NO = O + N_2$ are the major NO removal reactions at high temperatures. Studies show that the flame speed is extremely sensitive to the rate constant of the reaction $H + NO = N + OH$. The reactions involving HNO are found to be important in H_2-NO kinetics, the most sensitive reaction being the HNO decomposition reaction. Reactions involving N_2O make insignificant change in flame speed (less than 0.5%) at all conditions and can be deleted from the reaction set. A mechanism involving ten species (O_2, O, H_2, H, OH, H_2O, N_2, N, NO, HNO) and ten reversible reactions established here predicts flame speeds of H_2-NO system at various conditions of equivalence ratio, initial temperature, and pressure. Results are in good agreement with the experimental results of Magnus, Chintapalli, and Vanpee.

NOMENCLATURE

C_p	$\sum_{i=1}^{N_s} C_{pi} Y_i$, specific heat of the mixture at constant pressure [cal/(g)(K)]
C_{pi}	specific heat of ith species at constant pressure [cal/(g)(K)]
h_i	$(h_i^0 + h_i')$, sum of the sensible enthalpy and enthalpy of formation of the ith species (cal/g)
H	total enthalpy (cal/g)
N_s	number of species
P	pressure (atm)
R_a	gas constant = 1.9872 [cal/(mole(K)]
S_u	flame speed (cm/s)
T	absolute temperature (K)
t	time
X_i	mole fraction of species i
x	physical distance (cm)
Y_i	mass fraction of ith species
k	thermal conductivity of the mixture [cal/(cm)(s)(K)]

Greek Symbols

Δt	time step
ϕ	equivalence ratio
ρ	density of the mixture (g/cm^3)
ψ	distance coordinate = $\int \rho \, dx$ (g/cm^2)
τ	nondimensional temperature
$\dot{\omega}_i'''$	reaction rate of chemical species i [g/(cm^3)(s)]

Superscripts

$n + 1$	Intermediate time level
$-\infty$	Quantities at cold end
$+\infty$	Quantities at hot end

Subscripts

i	for species
u	for quantities at unburned end
b	for quantities at burned end
exp	measured experimentally

INTRODUCTION

The hydrogen-nitric oxide reaction, which involves an important reactant (nitric oxide) present in many combustion problems and also in atmospheric pollution, has attracted several researchers for about 60 years. The understanding of H_2-NO reaction mechanism is very important because the mechanism, involving the elements H, N, and O, forms the basis of more complex reaction systems such as monopropellant combustion of ammonium perchlorate.

Most of the early workers have studied the H_2-NO reaction at low temperatures (below 1500 K) [1-6]. During the 1970s, experimental as well

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as computational work on the H_2-NO reaction at very high temperatures (2000-4500 K) has been reported by several workers [7-11]. The major thrust during this time was to determine a rate constant for the reaction H + NO = N + OH, which is one of the important reactions in the extended Zeldovich mechanisms and also the rate-controlling step at high temperature. Rate constants for the reaction H + NO = N + OH as well as for the reaction NO + O = N + O_2 at relatively lower temperature are given by McCullough et al. [12]. Hanson and Salimian [13] have made an elaborate survey of rate constants for elementary reactions involving the species of elements nitrogen, hydrogen, and oxygen.

Bradley and Craggs [9] have reported studies on both complete and reduced mechanisms suggested by Wilde [6] by using up-to-date rate constants wherever possible. The predicted rates proved to be about three orders of magnitude below the experimental values. The discrepancy in the results is stated to be due to the significant uncertainty in the rate data of the dominant reaction HNO + NO = N_2O + OH, which was derived by Wilde. Criticizing Wilde's mechanism, Bradley and Craggs suggested three mechanisms for the H_2-NO reaction at high temperatures. Based on some arguments, only one mechanism containing eight species and five reactions was recommended for H_2-NO reaction kinetic modeling. Various other mechanisms of the H_2-NO reaction suggested by Flower et al. [7], Duxbury and Pratt [10], and Koshi et al. [11] are also investigated in the present article for the flame propagation problem.

Experimental studies on H_2-NO flames have been made by Parker and Wolfhard [14], Magnus et al. [15], Mainiero and Vanpee [16], and others. Although results on the H_2-NO flame by Parker and Wolfhard are cursory, Magnus et al. have given burning velocities of H_2-NO flames over the composition range of 35%-70% NO for two initial temperatures, 298 and 1023 K, at 1 atm. In addition, the burning velocity of the stoichiometric mixture (50% NO) is also given by them as a function of initial temperature over the temperature range of 300-1000 K. The bunsen burner method was employed by Magnus et al. [15] to perform the measurements. An averaged burning velocity was obtained from the measurement of the total volume flow of gas and the total area of the luminous flame front. The burner used by them was made up of a ceramic tube of 15 mm diameter with a ducted section of about 50 cm length. Mainiero and Vanpee [16] studied the reaction mechanism of the H_2-NO flame, stabilizing this flame on a Meker type burner in the temperature range of 2800-3100 K. Temperature profiles and concentration profiles of NO, OH, and O were determined in the reaction and postreaction zones using spectrophotometric techniques.

Computational studies on H_2-NO flames have not been reported so far. Even after a long research history, the reaction mechanism for H_2-NO flame problems has not been completely established, as it has for H_2-air and H_2-O_2 systems. There is a great deal of variability in the reaction mechanisms among different workers. The aim of the present work is to examine the sensitivity of H_2-NO flames to the rates of various elementary reactions and to establish a compact reaction mechanism for flame problems through careful literature survey and a comparison of the computational results with experiments.

THE GOVERNING EQUATIONS AND COMPUTATIONAL PROCEDURE

The conservation equations for species and energy in laboratory fixed coordinates describing a laminar premixed flame can be written as

$$\frac{\partial Y_i}{\partial t} = -\frac{\partial J_i}{\partial \psi} + \frac{\dot{\omega}_i'''}{\rho}, \tag{1}$$

$$\frac{\partial H}{\partial t} = \frac{\partial}{\partial \psi}\left[\frac{k\rho}{C_p}\frac{\partial H}{\partial \psi}\right] - \sum_{i=1}^{N_s}\frac{\partial}{\partial \psi}\left[\left\{J_i + \frac{k\rho}{C_p}\frac{\partial Y_i}{\partial \psi}\right\}h_i\right], \tag{2}$$

where ρ = mass density, k = thermal conductivity, C_p = specific heat of the mixture at constant pressure, H = total enthalpy of the mixture, and Y_i, h_i, J_i, and $\dot{\omega}_i'''$ are, respectively, the mass fraction, enthalpy, diffusive mass flux, and reaction rate of the ith species. The boundary conditions are

$$\text{as} \quad \psi\left(= \int \rho\, dx\right) \to -\infty; \qquad Y_i \to Y_{iu} \text{ and } T \to T_u, \quad (3)$$

$$\text{and as} \quad \psi \to +\infty; \qquad Y_i \to Y_{ib} \text{ and } T \to T_b. \quad (4)$$

Equations 1–4 are solved by using an operator-split method wherein reaction rate part and diffusion part are solved in two separate steps. In the first step, only the reaction rate part is solved by using an implicit scheme called linear block method [17] and the profiles of Y_i and T are obtained at the intermediate time level $n + 1$. These profiles of Y_i and T become the input for the diffusion part, which is solved using an explicit method for the same time step. Thus the complete computation for one step gives profiles of Y_i and T at the advanced time level $(n + 1)$. The flame speed is computed from these new profiles. Starting from an assumed initial profile (at $n = 0$ time level) for Y_i and T, the solution is time-marched until a steady state is reached. The details of the numerical scheme and its validation are reported in an earlier work [18]. The transport properties are calculated using the formulas taken from Brokaw's report [19] with the Lennard–Jones potential parameters given by Warnatz [20] and Svehla [21]. The reaction rates are calculated assuming all reactions to be reversible. The forward reaction rates are calculated using the law of mass action as applicable to multistep kinetics. The rates of the reverse reactions are computed from the forward rates and the equilibrium constants calculated from the thermodynamic data of the species, taken from Gordon and McBride [22].

RESULTS AND DISCUSSION

Investigation of Reaction Mechanism

In the study presented here, the species O_2, O, H_2, H, OH, H_2O, N_2, N, NO, N_2O, and HNO have been considered. The reactions containing species such as NO_2, HO_2, and NH etc. are not regarded as important in the literature for the H_2–NO reaction. The reactions involving NH, NH_2, etc., are important in systems involving ammonia, but in most of the early studies on shock tubes and flow reactors on H_2–NO systems, these species were found to be unimportant over a wide range of temperatures. The formation of the NH radical from the reactions involving HNO is unlikely because HNO is present in significant levels only at low temperature ($<$ 1800 K). The mechanisms containing the above mentioned species were proposed by Wilde [6], Bradley and Craggs [9], Duxbury and Pratt [10], Koshi et al. [11] and Flower et al. [7, 8]. Mechanisms suggested by Bradley and Craggs, Duxbury and Pratt and Koshi et al. do not contain N_2O. Rate constants of many reactions involving these species have been reviewed by Hanson and Salimian [13] and the rate constants proposed by them are shown in Table 1.

The flame propagation studies have been carried out using the mechanisms of various workers and the computational results are presented in Table 2 for stoichiometric mixture ($\phi = 1$). One can see from Table 2 that the flame speeds computed using various mechanisms vary widely from one another. The experimentally measured flame speed by Magnus et al. [15] for the same conditions is 227.5 cm/s. Only three mechanisms, namely the reduced mechanism of Bradley and Craggs [9] and the mechanisms of Flower et al. [7] and Hanson and Salimian [13], were found to produce a flame speed close to the experimental value. The final selection of the mechanism is based on the following considerations.

Bradley and Craggs reexamined Wilde's complete and simplified mechanisms with new rate constants for some of the reactions. They also felt the need for an additional mechanism, not contemplated by Wilde because the predicted rates (using Wilde's mechanism) proved to be about three orders of magnitude below their shock tube experimental results. A short reaction mechanism containing eight species and five reactions was proposed by them. The flame speed of the stoichiometric mixture computed by the present code using this mechanism is very close to the experimental value measured by Magnus et al. [15] (Table 2). However, this mechanism proved to be unsatisfactory for nonstoichiometric mixtures. For

TABLE 1

Elementary Reactions of the H_2-NO System [13] and Sensitivity of Flame Speed to Rate Constants[a]

No.	Reaction	A (cm^3/mol/s)	b	E (cal/mol)	% Variation in k	% Variation in S_u
R1	$H + NO = N + OH$	0.170×10^{15}	0.0	48801	$-40-+40$[b]	$-20-+11$
R2	$O + NO = N + O_2$	0.380×10^{10}	1.0	41369	$-30-+30$[b]	$-0.3-+0.2$
R3	$O + N_2 = N + NO$	0.182×10^{15}	0.0	76241	$-35-+35$[b]	$-3-+3$
R4	$HNO + M = NO + H + M$	0.178×10^{16}	0.0	48686	$-70-+70$	$-20-+25$
R5	$HNO + H = H_2 + NO$	0.126×10^{14}	0.0	4000	$-70-+70$	$-2-+2$
R6	$HNO + OH = H_2O + NO$	0.126×10^{13}	0.5	2000	$-70-+70$	$-1-+1$
R7	$HNO + O = NO + OH$	0.501×10^{12}	0.5	2000	$-70-+70$	$-2-+2$
R8	$N_2O + M = N_2 + O + M$	0.692×10^{24}	-2.5	64994	-100	< 0.5
R9	$N_2O + O = NO + NO$	0.692×10^{14}	0.0	26625	-100	< 0.5
R10	$N_2O + O = N_2 + O_2$	0.100×10^{15}	0.0	28016	-100	< 0.5
R11	$N_2O + N = N_2 + NO$	0.100×10^{14}	0.0	19870	-100	< 0.5
R12	$N_2O + H = N_2 + OH$	0.759×10^{14}	0.0	15101	-100	< 0.5
R13	$N_2O + O = N_2 + O_2$	0.100×10^{15}	0.0	28017	-100	< 0.5
R14	$N_2O + N = N_2 + NO$	0.100×10^{14}	0.0	19870	-100	< 0.5
R15	$NO + M = N + O + M$	0.398×10^{21}	-1.5	150000	-100	< 0.5
R16	$N_2 + M = N + N + M$	0.372×10^{22}	-1.6	224930		
R17	$H_2 + O = H + OH$	0.180×10^{11}	1.0	8900		
R18	$O_2 + H = O + OH$	0.220×10^{15}	0.0	16800		
R19	$H_2O + H = OH + H_2$	0.930×10^{14}	0.0	20360		
R20	$H_2O + O = OH + OH$	0.680×10^{14}	0.0	18360		
R21	$H_2 + M = H + H + M$	0.220×10^{14}	0.0	96000		
R22	$H_2O + M = H + OH + M$	0.130×10^{16}	0.0	105100		
R23	$O_2 + M = O + O + M$	0.355×10^{19}	-1.0	118000		

[a] Data for H-O reactions are taken from Ref. 8.

[b] Typical uncertainty quoted in Ref. 13.

example at $P = 1$ atm and $T_u = 1023$ K, the computed flame speeds were 128 cm/s for $\phi = 0.43$ (lean) and 225 cm/s for $\phi = 1.5$ (rich) compared with the measured values of 175 and 195 cm/s, respectively.

Reaction mechanisms proposed by Flower et al. [7], Duxbury and Pratt [10], and Koshi et al. [11] have many similar reactions and also the rate constants, but the difference in flame speeds predicted by these mechanisms is due to the difference in the rate constants of the reaction $H + NO = N + OH$. Wilde has not considered this reaction and on inclusion of this reaction in Wilde's reaction set, the predicted flame speed is obtained as 265 cm/s. The sensitivity of this reaction is further investigated in the next section.

TABLE 2

Computed Flame Speeds using Various Mechanisms
$P = 1$ atm, $\phi = 1$, $T_u = 1023$ K

Reaction Kinetic Mechanism for H_2-NO Reaction Suggested by	Flame Speed S_u Computed by Present Code (cm/s)[a]
1. Wilde [6] (Complete Set)	65
2. Bradley and Craggs [9]	227
3. Duxbury and Pratt [10]	315
4. Koshi et al. [11]	164
5. Flower et al. [7]	251
6. Hanson and Salimian [13]	243

[a] S_u(exp) = 227.5 cm/s [15].

The mechanisms proposed by Flower et al. [7] and Hanson and Salimian [13] gave flame speed within 10% of the experimental value for the stoichiometric mixture. Since Hanson and Salimian reviewed several data, including those of Flower et al. [7], the mechanism of Hanson and Salimian was taken up for further studies. As is shown later, this mechanism predicts consistently 7%-20% higher flame speeds compared with that of the experimental values at various mixture ratios and initial temperatures. The agreement is considered satisfactory at present since only one study is available for experimentally measured flame speed and hence the accuracy of measurement cannot be easily estimated. Also small changes in computed flame speeds are possible by the choice of transport properties.

Sensitivity of Reactions

Hanson and Salimian [13] have listed the rate constants for 16 reactions (excluding the hydrogen-oxygen reactions) involving the species mentioned in the previous section. Of these, seven reactions have been reviewed by them. The reactions of the extended Zeldovich mechanism and the reactions involving N_2O have been studied extensively in the literature. For the reactions involving HNO, the rate constants are based on scanty or low temperature data.

Zeldovich Reaction

Reactions R1-R3 of Table 1 are known as Zeldovich reactions and have been studied extensively in literature. Hanson and Salimian [13] have quoted the uncertainty factors for R1 between 0.6 and 1.4. The variation of rate constant within this range causes a change in flame speed in the range of −20% to +11%. The flame speed is fairly insensitive to the second reaction and almost no change in flame speed is observed by varying the rate constant by about 30%, which is the uncertainty range quoted for this reaction. The third reaction is moderately sensitive—about 3% change in flame speed is obtained by varying the rate constants by 35%.

Reactions Involving N_2O

The reactions involving N_2O are well studied, and reasonably accurate estimates of rate parameters are available. However the H_2-NO flame is quite insensitive to these reactions and even complete removal of these reactions does not change the flame structure. This species is not generated in any significant level during H_2-NO reaction (maximum value $\sim 0.5 \times 10^{-5}$). This conclusion is also consistent with earlier shock tube and flow reactor studies [7, 8, 12].

Reactions Involving HNO

The reactions involving HNO are R4-R7 in Table 1. No direct measurements of rate constants of these reactions are available, and the rate constants used are estimates. From Table 1 it can be seen that the flame speed is most sensitive to the rate constant of the HNO decomposition reaction. The sensitivity to this reaction is as high as that of the extended Zeldovich reaction R1 discussed earlier. The rate constants of the reactions involving HNO as reported by Hanson and Salimian [13] are about 70% lower than the values reported by Baulch et al. [23]. However, these reactions are not discussed in Ref. 13 and the source is quoted to be Baulch et al. [23].

It is interesting to note that the HNO decomposition reaction has not been found sensitive in shock tube [7, 8] and flow reactor [12] studies. The reactions involving HNO are important in the low temperature range. The maximum production rate of HNO is at a temperature of 1400 K and the maximum destruction rate is at 1700 K for the flame with initial temperature of 1023 K. Most of the shock tube studies are conducted at temperatures much above this range. However, it is surprising that even in flow reactor studies conducted by McCullough et al. [12] in the temperature 1750-2400 K this reaction was found very insensitive.

Compact Reaction Set for H_2-NO Flame

The sensitivity analyses of the previous section have shown that the flame speed is very insensitive to some of the reactions and a compact reaction set can be found that would be satisfactory for computing H_2-NO flames. In order to find out whether the mechanism shown in Table 1 can be made shorter by reducing species and reactions, the following five groups are made from this mechanism: (1) N_2O-containing reactions, (2) HNO-containing reactions, (3) O_2-containing reactions, (4) dissociation reactions except dissociation of HNO (R16, R21, R22, and R23), and (s) the reactions involving species other than N_2O, HNO, and O_2. The species other than N_2O, HNO, and O_2 are O, H_2, H, OH, H_2O, N_2, N, and NO. Of these eight species, six (O, H, OH, N_2, N, NO) are involved in two important reactions of the extended Zeldovich mechanism ($H + NO = N + OH$, $O + N_2 = N + NO$) and the other two species are as reactant (H_2) and product (H_2O). Therefore, no attempt is made to neglect the reactions and species of group 5. The effect of neglecting the reactions of the other four groups, either singly or in conjunction with others, has been examined (Table 3). Neglect of HNO-containing reactions leads to a

TABLE 3

Effect of Neglecting Some Species and Reactions of Table 1 on Flame Speed, $P = 1$ atm, $\phi = 1$, $T_u = 1023$ K, $S_u(\mathrm{exp}) = 227.5$ cm/s

Sl. No.	Reaction Mechanism	Flame speed (cm/s)	% diff
1.	All the reactions of Table 1 [denoted as Full Set (FS)]	243.0	0.0
2.	FS minus (−) N_2O-containing reactions	242.5	−0.4
3.	FS − HNO-containing reactions	181.0	−25.0
4.	FS − O_2-containing reactions	242.0	−0.2
5.	FS − N_2O dissociation reactions (R16, R21, R22, R23)	242.0	−0.2
6.	FS − N_2O-containing reactions and also R16, R21, R22, R23	241.5	−0.6
7.	FS − N_2O- and O_2-containing reactions and also R10, R11, R12	241.5	−0.6
8.	FS − HNO-containing reactions and also R16, R21, R22, R23	No convergence	—
9.	FS − N_2O- and HNO-containing reactions	181.0	−25.0

significant change in flame speed (c ase 3 in Table 3) while neglect of N_2O-containing or O_2-containing reaction s results in a small amount of change in flame speed (cases 2 and 4 in Table 3, respectively).

Removal of dissociation reactions (R16, R21, R22, and R23) from the full set causes a very small change in flame speed (case 5 in Table 3) but if these are removed along with HNO-containing reactions the steady state is not attained (case 8 in Table 3). Of the four dissociation reactions, $H_2 + M = H + H + M$ is the main reaction for chain initiation when taken in absence of HNO-containing reactions [9]. On the other hand, it is observed in the present analysis that in the lower temperature range, HNO-containing reactions are very important. This is indeed confirmed from Fig. 1 where rection rates ($\dot{\omega}_i'''/\rho$) of various species are plotted against nondimensional temperature. It can be seen from Fig. 1 that, while most species have peak reaction rates close to flame temperature (~ 3000 K), HNO has peaks (both positive and negative) below 2000 K. The reaction rate of HNO decays slowly towards the hot boundary. When HNO-containing reactions are present in the mechanism, the presence or absence of dissociation reactions does not make much of a difference (cases 1, 2, 4, 5, 6, and 7 in Table 3). This is found to be true in all conditions ranging from lean to rich and from lower to higher pressures. Hence it is possible to remove the earlier mentioned dissociation reactions from the full set.

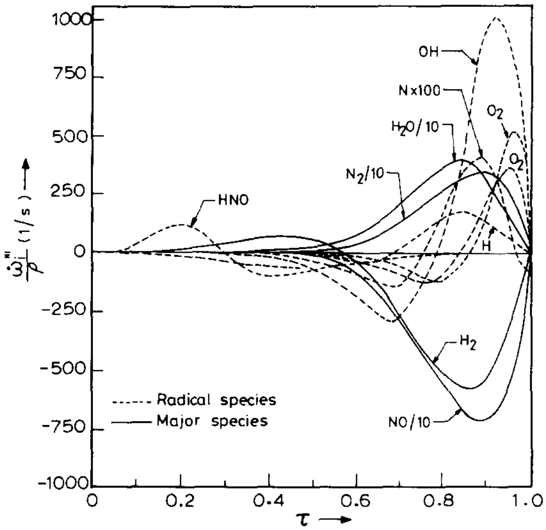

Fig. 1. Variation of reaction rates of different species with nondimensional temperature τ ($T_u = 1023$ K, $P = 1$ atm, $\phi = 1$)

It is interesting to note that the removal of O_2-containing reactions in the stoichiometric flame does not produce much change in flame speed (case 4 in Table 3). On the other hand, these reactions are very important in lean flames since computation with a mechanism excluding these reactions does not even converge. The removal of N_2O-containing reactions, however, makes very little change in all the conditions.

Finally, removal of the dissociation reactions and N_2O-containing reactions yields a reduced set containing ten species and ten reversible reactions. Table 4 gives the reduced set of reactions and rate constants. The validity of this set has been examined by comparing the computational results with experimental results over a wide range of temperature and composition. These results are presented in the next section.

Detailed Comparison with Experimental Results

In this section the flames computed using the reduced set of reactions obtained in the previous section are compared with the experiments. In

TABLE 4

Compact Reaction Set for Modeling H_2-NO Flame

No	Reaction	A (cm^3/mol/s)	b	E (cal /mol)
1	$H + NO = N + OH$	0.170×10^{15}	0.0	48801
2	$O + NO = N + O_2$	0.380×10^{10}	1.0	41369
3	$O + N_2 = N + NO$	0.182×10^{15}	0.0	76241
4	$HNO + M = NO + H + M$	0.178×10^{16}	0.0	48686
5	$HNO + H = H_2 + NO$	0.126×10^{14}	0.0	4000
6	$HNO + OH = H_2O + NO$	0.126×10^{13}	0.5	2000
7	$H_2 + O = H + OH$	0.180×10^{11}	1.0	8900
8	$O_2 + H = O + OH$	0.220×10^{15}	0.0	16800
9	$H_2O + H = OH + H_2$	0.930×10^{14}	0.0	20360
10	$H_2O + O = OH + OH$	0.680×10^{14}	0.0	18360

Fig. 2, the computed profiles of temperature and mole fractions of NO, OH, and O are compared with the experimental measurements of Mainiero and Vanpee [16]. The agreement seems to be reasonable considering the fact that the computations were made for adiabatic conditions while there was heat loss in the experiment, as evident by the fall in temperature beyond the flame zone (Fig. 2). Because of the fall in temperature, concentration also changes due to the shifting equilibrium. The comparison between computations and experiments is reasonable in the early part of the flame, but becomes poor beyond that.

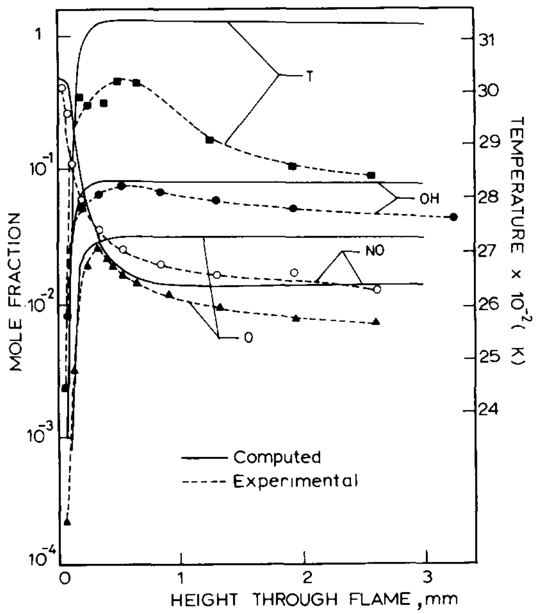

Fig. 2. Profiles of temperature and mole fractions of NO, O, and OH. Comparison of present results with experimental measurements of Mainiero and Vanpee [16] ($P = 1$ atm, $\phi = 1$, $T_u = 298$ K).

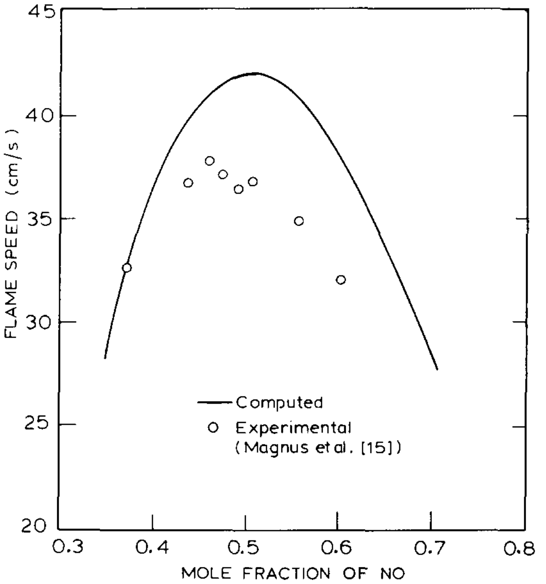

Fig. 3. Variation of S_u with mole fraction of NO at $T_u = 298$ K and $P = 1$ atm. Comparison of the present results with experiments of Magnus et al. [15].

Figures 3 and 4 show the variation of flame speed with mixture ratio at initial temperature 298 and 1023 K, respectively. The computed flame speeds agree better at 1023 K (~ 10% higher than measured values) than at 298 K

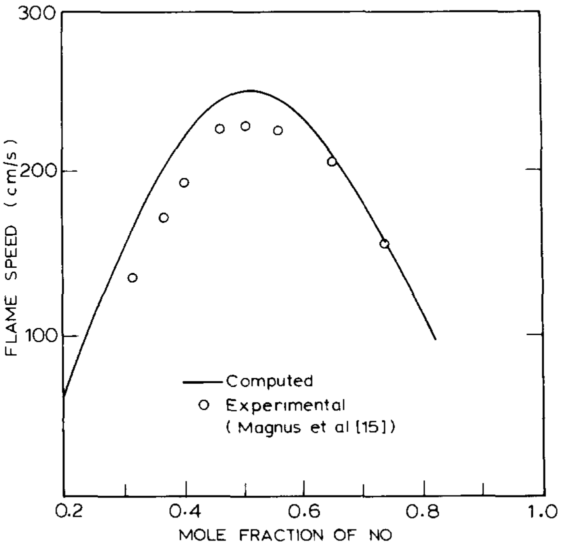

Fig. 4. Variation of S_u with Mole Fraction of NO at $T_u = 1023$ K and $P = 1$ atm. Comparison of the present results with experiments of Magnus et al. [15].

(~ 18% higher than measured values). The reason for this may be attributed perhaps to the accuracy of the measurement in view of the difficulty in stabilizing the flame at $T_u = 298$ K [15].

The flame speeds have also been computed at various initial temperatures, and the results are plotted in Fig. 5 for stoichiometric mixture. The computed flame speeds are consistently higher than the experimental values, the agreement becoming better as the temperature is increased.

Some computations were also made to obtain the effect of pressure on flame speed and the results are shown in Fig. 6. The rate constants of the decomposition/recombination reaction could be pressure sensitive at high pressures. However, no reliable falloff data are available for the falloff behavior of these reactions. The falloff behavior seems less important for simple molecules like H, O, and OH. Hence the falloff behavior is not considered in the present calculations. No experimental results are available to compare with the computations but the increase in flame speed with pressure in the subatmospheric range seems to be consistent with the results of the H_2-N_2O system of Parker and Wolfhard [14]. The increase of the flame speed in the low-pressure range may be explained by the increase in flame temperature with pressure. The flame temperatures of stoichiometric mixtures of H_2 and NO at an initial temperature of 1023 K are 2634, 2903, 3216, 3575, 3974, and 4389 K at pressures of 0.01, 0.1, 1, 10, 100, and 1000 atm, respectively. Since the peak reaction rates of most of the species occurs at temperatures close to the flame temperature, substantial increase in the reaction rate is possible with the increased flame temperature. The overall order of reaction varies from 2.1 below 1.0 atm to 1.8 beyond 10 atm.

CONCLUSIONS

The H_2-NO flame has been studied computationally and the sensitivity of important reactions has been examined. A reaction set containing ten species and ten reversible reactions was found to be adequate to describe H_2-NO kinetics quite accurately. This has been demonstrated by com-

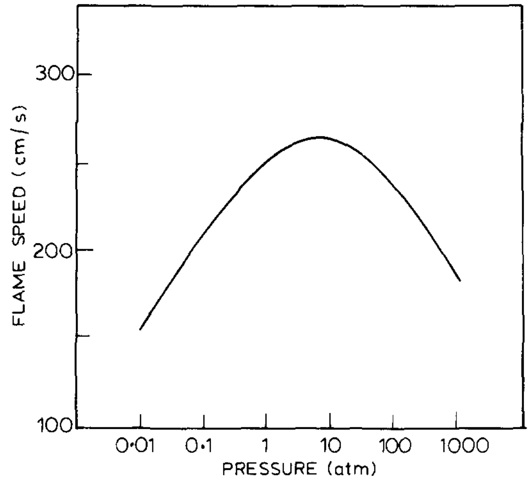

Fig. 6. Variation of computed flame speed with pressure at $\phi = 1$ and $T_u = 1023$ K.

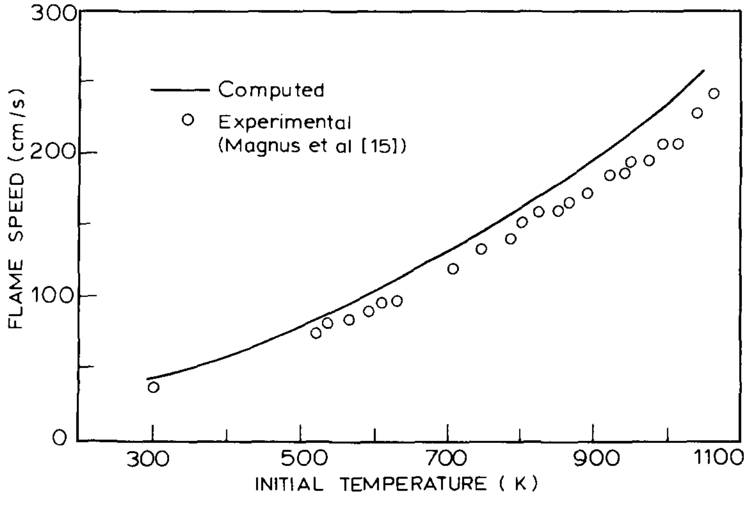

Fig. 5. Variation of S_u with initial temperature at $\phi = 1$ and $P = 1$ atm. Comparison of the present results with experiments of Magnus et al. [15]

paring the computed results with experiments over a wide range of conditions. The main conclusions from the study are as follows:

1. At higher temperatures, the reaction NO + H = N + OH is responsible for NO removal. The computed flame speed is very sensitive to the rate data of this reaction.
2. Reactions involving HNO were found to be important in H_2–NO reaction mechanism. These reactions on exclusion make significant change in flame speed. The most sensitive reaction in this group is the HNO decomposition reaction.
3. N_2O is unimportant in the H_2–NO reaction mechanism.
4. The computed flame speeds at various flame conditions are in reasonably good agreement with the burning velocities measured by Magnus et al. [15]. Also the computed profiles of T, OH, NO, and O are in qualitative agreement (in flame zone) with those measured by Mainiero and Vanpee [16].

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