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# Structure and chemical kinetics of flames supported by nitrogen oxides*

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ABSTRACT

This paper is a report on efforts to model burner-stabilized and free-standing flames supported by nitrogen oxides. A 272-reaction mechanism has been used as a basis for modeling flames supported by the oxidizers $N_2O$, $NO_2$ and NO. Results have been compared with recent experimental data for burner-stabilized flames and free-standing flames. For burner-stabilized flames, comparisons between modeling and experimental data have been made for major species and radical concentrations. Where discrepancies between modeling and the data exist, analysis has been performed to identify key mechanisms contributing to species production rates in the flame. For free-standing flames, comparisons between modeling and experimental data have been made for flame velocities. Analysis has also been performed to identify key mechanisms in these flames. In general, the modeling results have been found to predict accurately major species concentrations in burner stabilized flames. Problems have been encountered, however, in accurately modeling the qualitative behavior of radical species. Reasonable agreement has been found between the modeling and experimental data for freely-propagating flames.

## INTRODUCTION

The combustion of solid rocket propellants and other energetic materials is a complex multidimensional and multiphase process involving a wide variety of chemical species. The very high pressure and temperature conditions of practical rocket combustion chambers are at present inaccessible by most conventional diagnostic techniques. The study of these coupled phenomena in situ, therefore, has not been possible in sufficient detail to develop a complete understanding of the chemistry and physics of the combustion process. The objective of most recent studies of the combustion of these materials has been to study separate aspects of the overall process in an effort to provide a comprehensive understanding of the combustion mechanism. This study is one component of that coordinated investigation and has as its focus the gas phase reactions associated with the combustion of these solid fuels.

The decomposition of many of these solid energetic materials during combustion leads to the formation of gaseous hydrocarbon fuel species and oxides of nitrogen which serve as oxidizers (ref. 1 and 2). The reactions of these decomposition products above the propellant surface lead to a gaseous flame which can provide heat which is transferred back to the propellant surface and can thereby influence the burning rate. The purpose of this paper is to summarize the current status of studies we have undertaken of model gas phase flames associated with the combustion of nitramine bases solid rocket propellants. These studies consist of measurements of the structure of stable and unstable species concentration profiles and temperature in laminar, premixed, flat flames of fuel/$NO_x$ mixtures at low pressure. The experimental measurements are then compared to calculations of the concentration profiles using a one dimensional flame code which models the transport processes and chemistry of the flame. The transport processes include species diffusion and thermal conduction through the flame and the chemistry is modeled by a detailed chemical kinetic reaction mechanism.

The basic mechanism used for the modeling work is a subset of the 331-reaction mechanism of Volponi and Branch (ref. 3). Reactions removed were those involving species with low concentrations, including condensed phase molecules, and molecules containing more than three carbon atoms. The resulting mechanism contains 272 reactions. This mechanism was the basis for all of the modeling described in this paper. The flame code used to solve the one dimensional flame equations was by Kee et al. (ref. 4)

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## FLAMES SUPPORTED BY $N_2O$ AS OXIDIZER

Several recent studies have focused on measuring the temperatures and species concentrations in flames supported by $N_2O$. Habeebullah et al. (ref. 5), Zabarnick (ref.6), and Vandooren et al. (ref. 7) studied $CH_4$ reacting with $N_2O$. Habeebullah measured stable species concentrations using probe sampling, radical species concentrations using laser-induced fluorescence (LIF), and flame temperatures using thermocouples and LIF. Zabarnick used LIF to measure stable and radical species concentrations and flame temperatures. Dindi et al. (ref. 8) studied CO reacting at $N_2O$, and used probe sampling to measure stable species concentrations, LIF to measure unstable species concentrations, and thermocouples to measure flame temperatures.

We have modeled the structure of all of these flames using our 272 step reaction mechanism. It was found that in order to have best agreement with the full range of data represented by these results, the rates of several of the reactions needed to be adjusted as indicated in Table 1. The mostsignificant of these changes was for the $N_2O$ decomposition which was adjusted to within the upper range of the literature data for this reaction. This change was needed to model the methane flame data correctly. The other reactions listed in Table I were adjusted to provide good agreement for the $CO/N_2O$ flame data. The results of the modeling for the most important cases studied are given below for the flames with $N_2O$ as oxidizer.

### CO–$N_2O$ flame structure

Low pressure, laminar, premixed flames of CO-$N_2O$ have been stabilized over a rectangular flat-flame burner (ref. 8). Laser-induced fluorescence spectroscopy was used to establish the absence of CN, CH, NH, $NH_2$ and OH in these flames. Gas chromatographic sample analysis was used to determine the CO, $CO_2$, $N_2O$, NO, $N_2$ and $O_2$ concentration profiles for three CO-$N_2O$ flames having equivalent ratios of 1.00, 1.32, 1.50. Lean flames could not be stabilized. Temperature profiles for all three flames were measured using R type thermocouples. Measured temperature profiles were corrected for radiation losses. These flames are considerably lifted above the burner and contain a single luminous zone. The main feature of these flames is the absence of any reactive intermediates except oxygen atom. We have eliminated reactions involving the element H and reduced the 272 step mechanism to 27 reactions. This mechanism was then used to model these CO-$N_2O$ flame data.

A combined "elementary reaction contribution" and "sensitivity" analysis showed that only four of the 27 elementary reactions used in the original kinetic mechanism were important in the production or the consumption of major species in the flame. A comparison of the calculated concentration profiles using the original 27-reaction and the four-reaction mechanisms for all three CO-$N_2O$ flames studied in this investigation showed that the difference between the two profiles was always less than 1% of the total concentration. Hence we propose the following four-step mechanism for the CO-$N_2O$ flames:

$$CO + N_2O \rightarrow CO_2 + N_2$$
$$N_2O + M \rightarrow N_2 + O + M$$
$$N_2O + O \rightarrow N_2 + O_2$$
$$N_2O + O \rightarrow NO + NO$$

The first of the four reactions listed above is the most important reaction for CO-$N_2O$ flames. It accounts for almost all the CO consumption and nearly 90% of the $N_2O$ consumption. The second reaction ($N_2O+M \rightarrow N_2+O+M$) is an important reaction for kineticists. It plays a key role in various environments for $NO_x$ formation or decomposition. It is also used for generating O atoms in studies of elementary oxidation reactions. The last two reactions of the four-step mechanism listed above control the concentration profiles of $O_2$ and NO.

### $CH_4$–$N_2O$ flame structure

Laminar, premixed flat flames of $CH_4$ with $N_2O$ have been stabilized and studied at 50 torr (ref. 5). This study represents the first nearly complete study of the structure and kinetics of $CH_4$-$N_2O$ flames including stable and unstable species measurements and detailed chemical kinetic modeling. Three flames were investigated, with slightly fuel rich, near stoichiometric and lean mixtures. Flame modeling has been done using the 272 step reaction mechanism and the reaction mechanism evaluated.

The results of the flame calculations and the experiments are given in Figure 1. The comparison shows good quantitative and qualitative agreement between the measured and calculated profiles especially for stable species. The modeling simulation clearly predicts the general flame structure and species concentration profiles. For the flame radicals, the theoretical results are in good agreement with the experimental results for NH, CN and OH. The computed maximum for the CH radical is shifted in the flame. The exact prediction of the radical concentrations using flame modeling is difficult owing to the high reactivity of these radicals, especially CH.

The reaction mechanism starts with the thermal decomposition of $N_2O$ to $N_2$ and O which is the initial reaction for radical formation in the flame. The oxygen atom formed from this reaction reacts with $H_2$ to form a pool of H and OH radicals which then propagate the rest of the mechanism. The results of the sensitivity analysis

TABLE 1. Rate constants used in the present 272 Step Reaction Mechanism which differ from those of Volponi and Branch (Ref. 3). Units are mole, $cm^3$, sec, K, and Cal/mole.

| Reaction | A | n | Ea |
|---|---|---|---|
| $N_2O+OH=N_2+HO_2$ | 1.00e13 | 0 | 10000 |
| $N_2O+H=N_2+OH$ | 1.50e14 | 0 | 15090 |
| $N_2O+M+N_2+O+M$ | 4.90e15 | 0 | 57500 |
| $N_2O+O=N_2+O_2$ | 7.00e14 | 0 | 28200 |
| $N_2O+O=2NO$ | 5.60e14 | 0 | 28200 |
| $CO+N_2O=CO_2+NO$ | 2.00e12 | 0 | 17300 |

TABLE 2. Comparison of measured and calculated flame speed of fuel/$NO_x$ mixtures[a].

| REACTANTS | MEASURED[b] | CALCULATED | |
|---|---|---|---|
| | | M/B[c] | This Work[d] |
| $H_2/N_2O$ | 300 | 227 | 240 |
| $CH_4/N_2O$ | 105 | 76.2 | 90 |
| $C_2H_2/N_2O$ | 160 | 128 | 150 |
| $C_2H_4/N_2O$ | 110 | 94 | 102 |
| $C_2H_4/NO_2$ | 62.5 | 61 | 61 |

(a) All flame speeds given in cm/sec at 0.10 atm and for a stoichiometric mixture.
(b) Parker & Wolfhard (Ref. 12).
(c) Calculation using mechanism of Miller & Bowman (Ref. 13).
(d) Calculation using the present 272 step mechanism.

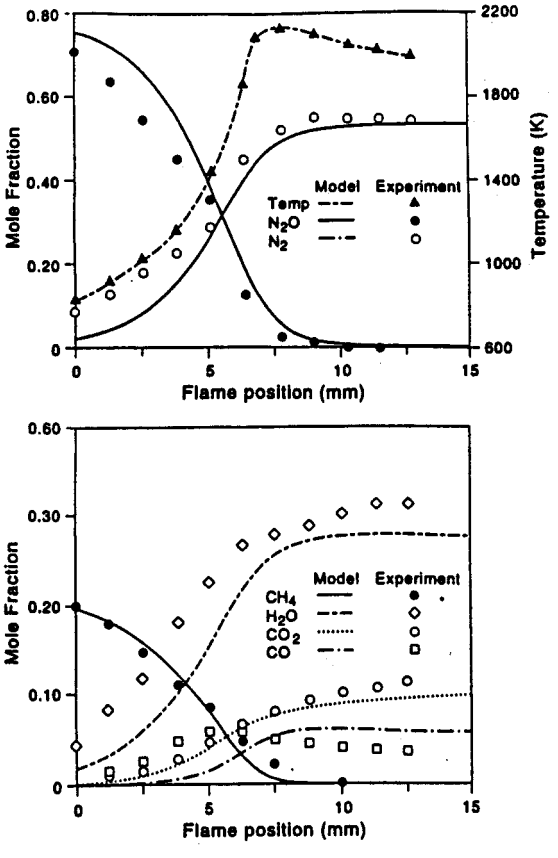


Figure 1. Comparison of $CH_4/N_2O$ flame structure data of Habeebullah et al. (Ref. 5) to calculations of flame structure using 272 step reaction mechanism.

also shows that $N_2O$ is not totally decomposed to $N_2$, but it forms some NH and NO molecules directly through reaction with H atoms. The mechanism also shows that methane has a long induction period before being consumed mainly through radical reactions (especially with OH and H) to form methyl ($CH_3$) intermediate. Once methyl intermediate is formed it has two reaction paths to follow. The first path is to form formaldehyde ($CH_2O$) which then starts a reaction path to form $CO_2$. The major reaction for CO conversion to $CO_2$ in this path is through its reaction with OH radical. The second path that $CH_3$ follows is to form $CH_2$ intermediate which then forms CH through radical reactions. The CH formed through this path reacts with NO from $N_2O$ decomposition to produce NH or CN radicals. This reaction forms the first link in hydrocarbon-nitrogen interactions. The high CN and NH concentration in the flame is attributed to this reaction. However, the reaction of nitrous oxide ($N_2O$) with H atoms is another important source for the formation of these two radicals (CN and NH). The final products in the exhaust gas were mainly $N_2$, $H_2O$, CO and $CO_2$ with small amounts of NO.

## FLAMES SUPPORTED BY $NO_2$ AS OXIDIZER

Five flames supported by $NO_2$ as the oxidizer have been studied recently and were tested against our 272 step reaction mechanism. Volponi and Branch (ref. 3 and 9) studied $H_2$ and $C_2H_2$ reacting with $NO_2$ in an argon diluent. They measured stable species concentrations using probe sampling, radical species concentrations using LIF, and flame temperatures using thermocouples and LIF. Branch et al. (ref. 10) studied two different flames supported by $NO_2$, $CH_4/NO_2/O_2$ and $CH_2O/NO_2/O_2$. They measured stable species concentrations using probe sampling, unstable species concentrations using LIF, and flame temperatures with a thermocouple. Zabarnick (ref. 11) also studied the $CH_4/NO_2/O_2$ flame, using LIF to measure flame temperatures and stable and radical species concentrations. The results of our current modeling of the hydrogen and the acetylene flames are most indicative of the reaction mechanisms involved and are discussed below.

### $H_2$–$NO_2$ flame structure

Measurements of the composition of stable and unstable species and temperature in laminar, premixed, flat flames of $H_2$-$NO_2$-Ar have been made and compared to the structure calculated with a flame code including detailed chemical kinetics (ref. 9). No previous detailed flame structure measurements and chemical kinetic modeling of this flame have been presented. Similar measurements and calculations are reported for a companion $H_2$-$O_2$-Ar flame in order to provide a comparison to a previously well characterized flame. We have modeled these flame measurements using the 272 step reaction mechanism with the carbon species removed. The resulting 87 step mechanism gives good agreement to the experimental data as shown in Figure 2.

In contrast to the $H_2O$-$O_2$-Ar flame, the rate and sensitivity calculations for $H_2$, $NO_2$, $H_2O$ and OH in the $H_2$-$NO_2$-Ar flame show that species net reaction rates are usually dominated by a single reaction. Almost all of the $H_2$ consumption and $H_2O$ formation is by $OH+H_2=H_2O+H$. The consumption of $NO_2$ and formation of NO is almost entirely by $NO_2+H=NO+OH$. The sum of these two reactions gives the global reaction of the flame $H_2+NO_2=NO+H_2O$. There is some formation of $O_2$ early in the flame by $NO_2+O=NO+O_2$ followed by $O_2$ consumption by $O_2+H=O+OH$. The OH is a balance of formation by $NO_2+H=NO+OH$ and consumption by $OH+H_2=H_2O+H$.

The reactions with greatest negative sensitivity for $H_2$ and $NO_2$ have the highest positive sensitivity for NO and $H_2O$. The reactions with the largest positive sensitivity for $H_2$ and $NO_2$ likewise have the largest negative sensitivity for NO and $H_2O$. The OH is primarily sensitive to its major formation and consumption reactions.

The importance of the reaction $H_2+NO_2=HNO_2+H$ to the $H_2$-$NO_2$-Ar flame mechanism is observed in the sensitivity and analysis for $H_2$, $NO_2$, NO, $H_2O$ and OH. This reaction provides the most important initiation step. It also has a significant effect on the OH profile by producing H early in the flame and promoting the formation of OH by $NO_2+H=NO+OH$. The $HNO_2$ is subsequently consumed by several reactions. Using a mechanism that contains only the three reactions

$$NO_2 + H = NO + OH$$
$$OH + H_2 = H_2O + H$$
$$H_2 + NO_2 = HNO_2 + H$$

gives a flame model that compares to within 5% of the model using the entire reaction mechanism.

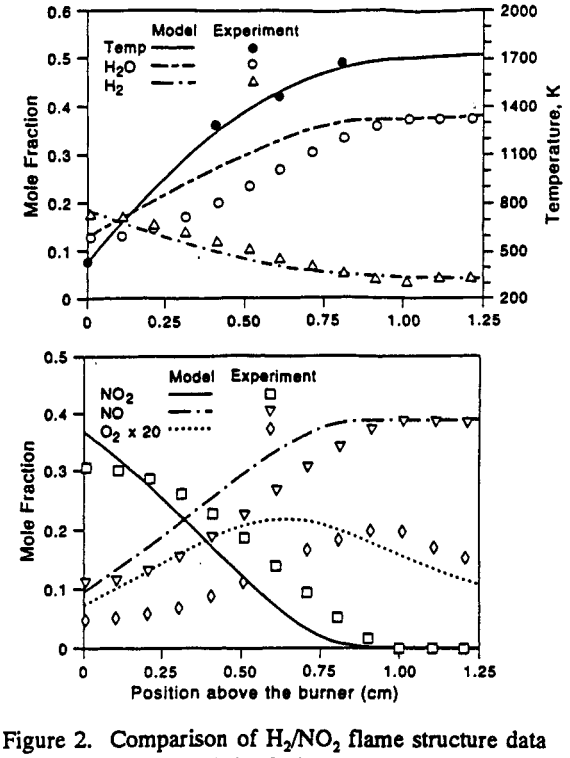


Figure 2. Comparison of $H_2/NO_2$ flame structure data of Volponi and Branch (Ref. 9) to calculations of Flame Structure Using 272 step reaction mechanism.

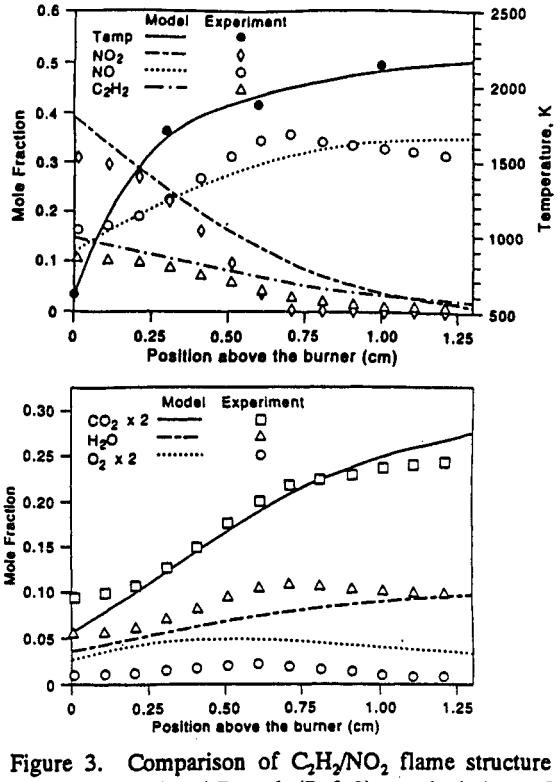


Figure 3. Comparison of $C_2H_2/NO_2$ flame structure data of Volponi and Branch (Ref. 3) to calculations of flame structure using 272 step reaction mechanism.

### $C_2H_2$–$NO_2$ flame structure

Volponi and Branch (ref. 9) studied $C_2H_2$ reacting with $NO_2$ in an argon diluent at a pressure of 25 torr. We modeled this flame using the 272 step reaction mechanism and the measured temperature profile. The modeling results using this mechanism are close to those Volponi and Branch obtained using the full, 331-reaction mechanism from which the 272-reaction mechanism was derived. This indicates that modeling results are not significantly affected using the smaller mechanism.

Measured and modeled mole fraction profiles for major species are shown in Figure 3. Discrepancies can be seen near the burner surface, as the modeling overpredicts the surface mole fractions of unreacted species and underpredicts the surface mole fraction of product species. Agreement is good, however, in the general trends shown in the profiles, and in the final mole fractions obtained in the flame. The modeling was used to find the principal reactions involved in each specie's production/consumption. The principal reaction consuming $C_2H_2$ is $C_2H_2+OH=C_2H+H_2O$; the principal reaction consuming $NO_2$ is $NO_2+H=NO+OH$; the principal reaction producing NO is $NO_2+H=NO+OH$; the principal reaction producing $O_2$ is $2NO_2=2NO+O_2$; the principal reaction producing $CO_2$ is $NO_2+CO=NO+CO_2$, and the principal reaction producing $H_2O$ is $OH+C_2H_2=C_2H+H_2O$. The importance of radicals in breaking down the unreacted species is evident.

Comparisons were also made between modeled and measured mole fractions for the radical species OH and CN. There were discrepancies in the calculated and measured trends for these species. The principal reaction involving OH production is $NO_2+N=NO+OH$, and the principal reaction involving CN production is $HCN+OH=CN+H_2O$, and the principal reaction involving CN production is $HCN+OH=CN+H_2O$.

Volponi and Branch (ref. 3) also modeled $C_2H_2$ reacting with $O_2$ with Argon gas as a diluent at a pressure of 25 torr. We modeled this flame using the 272 step mechanism and the measured temperature profile. The modeling's accuracy was on a par with what was obtained for the $CH_4/N_2O$ flame. Modeling was generally in good agreement with experimental data for major species concentrations in the flame, with the exception of $H_2O$, whose concentration was slightly underpredicted. Modeling and experiments agreed in the general trends for the concentration of radical species OH and CH, but modeling overpredicted the peak concentrations of both species by a factor of two. As with the $CH_4/NO_2$ flame, our modeling results compared well with those Volponi and Branch obtained using the 331-reaction mechanism.

## FLAMES SUPPORTED BY NO AS OXIDIZER

Zabarnick (ref. 6) studied a $CH_4/NO/O_2$ flame at 63 torr, using LIF to measure temperature and stable and radical species concentrations. This flame was modeled using the measured temperature profile and the 272 step mechanism.

The measured NO concentration profile showed a more pronounced drop-off to a steady-state value than modeling predicted. Also, the final calculated value of NO in the flame is 50% higher than what was measured; NO was the only stable species Zabarnick performed measurements on in this work.

Modeling and experimental results were compared for radical species. The predicted peak concentration of CN leads the experimentally measured peak by about 1 mm. The predicted OH concentration showed a drop-off late in the flame that was not detected experimentally. The predicted NH peak led the measured peak by about .5 mm. The predicted CH peak led the measured peak by about 1 mm also. Note that modeling now leads the experimental data for the location of peak radical concentrations, while for the $CH_4/N_2O$ flame also measured by Zabarnick (ref. 6) modeling predictions generally lagged the experimental data.

## *BURNING VELOCITY OF FUEL/$NO_x$ LAMINAR PREMIXED FLAMES*

The 272 step reaction mechanism was used to model the burning of free-standing flames supported by $N_2O$ and $NO_2$, and the modeled flame speed was compared to the experimental results obtained by Parker and Wolfhard (ref. 12) using the bunsen burner technique (Table 2). Also shown are the burning velocities obtained with the Miller and Bowman (ref.13) mechanism without the modifications discussed in the present investigation. The results indicate that generally good agreement is obtained between the modeling and the experimental data and that the 272 step mechanism gives better results than earlier mechanisms. The burning velocity of the flames with $N_2O$ are significantly higher than those with $NO_2$ as oxidizer. Two of the calculations are discussed below in more detail in order to indicate the importance of fuel and oxidizer decomposition reactions in flame propagation.

Table 1 gives the calculated and measured results for the burning rate of a $CH_4/N_2O$ flame. The calculated flame velocity is 90 cm/sec, very close to the measured velocity of 105 cm/sec. In addition, for this flame we were able to perform analysis on what reactions contribute most strongly to a given specie's production/consumption. It was found that the major reactions leading to the decomposition of $CH_4$ are the following, in decreasing order of their rate of $CH_4$ consumption:

$$CH_4 + H = CH_3 + H_2$$
$$CH_4 + OH = CH_3 + H_2O$$
$$CH_4 + O = CH_3 + OH$$

The major reactions leading to the breaking down of $N_2O$ were found to be, in decreasing order of the amount of $N_2O$ they consume:

$$N_2O + H = H_2 + OH$$
$$N_2O + M = N_2O + M$$

The importance of the radicals H, OH and O are clearly evident in the propagation of this flame.

A free-standing $C_2H_4$ flame at a pressure of 0.1 atm was modeled and the calculated flame speed was compared to that measured by Parker and Wolfhard (ref. 10). The results are included in Table 2, and as can be seen, the agreement is excellent for this flame. It was difficult to obtain a converged solution modeling many $NO_2$-supported flames, so there are not more cases reported at this time. It is possible that the larger mechanism used to model this flame aided in obtaining better agreement with experimental data than was obtained in several of the cases of $N_2O$-supported flames. More cases would need to be run, however, to draw conclusions on the relative ability of the two mechanisms to model free-standing flames supported by $NO_2$ and $N_2O$.

Modeling results for this flame showed that the top four reactions contributing to the decomposition of $C_2H_4$ in the flame were the following:

$$C_2H_4 + OH = C_2H_3 + H_2O$$
$$C_2H_4 + M = C_2H_2 + H_2 + M$$
$$C_2H_4 + H = C_2H_3 + H_2$$
$$C_2H_4 + O = CH_3 + HCO$$

The top four reactions leading to the decomposition of $NO_2$ were found to be:

$$NO_2 + N = NO + OH$$
$$NO_2 + CO = NO + CO_2$$
$$NO_2 + M = NO + O + M$$
$$NO_2 + OH = HO_2 + NO$$

Initiation of the chemistry in these flames can be seen to be closely tied to reactions producing radical species.

## SUMMARY AND CONCLUSIONS

We have completed detailed comparisons of calculations and measurements of the structure and burning velocity of fuel/$NO_x$ mixtures using a 272 step reaction mechanism. The comparison between the calculated flame structure and the experimental flame structure for stable species was found to be very good for fuel/$N_2O$ flames and good for fuel/$NO_2$ flames. The concentration profiles for radical species were found to be generally well represented qualitatively but not well represented quantitatively. It was concluded that, despite some remaining difficulties with the reaction mechanism, it appeared to be reliable in describing the overall combustion behavior of a wide range of fuel and oxidizer mixtures.

The most important reactions of the oxidizer ($N_2O$ or $NO_2$) are with H atoms and, to a lesser extent, with CO. Reaction of either oxidizer with H is a chain propagating reaction, in contrast with the chain branching reaction of H with $O_2$ which is of equal importance in fuel oxidation by $O_2$. In addition, the reaction of $NO_2$ with H is slower than $N_2O$ with H. Finally, the reaction of $N_2O$ with CO can be significant both in consumption of CO and formation of $CO_2$. This situation is again in contrast with the oxidation chemistry of systems by $O_2$ in which the conversion of CO to $CO_2$ is almost entirely be reaction of CO with OH. The difference between the use of $N_2O$ or $NO_2$ as oxidizer is that the former produces primarily $N_2$ while the latter produces NO. The subsequent

slow reduction of NO to $N_2$, even when it is thermodynamically favored, accounts for the most striking difference between the two oxidizers. The most important effects of the oxidizer, therefore, are that the nitrogen oxides are less effective chain carriers and lead to slower reaction rates compared to $O_2$.

The oxidation reactions of $CH_4$ in the presence of $NO_x$ is generally similar to the oxidation by $O_2$. Chain propagating and branching reactions lead to the formation of H, O and OH and these species progressively abstract hydrogen and partially oxidize $CH_3$ to $CH_2O$. The $CH_2O$ is then converted largely to CO through the intermediate HCO. Subsequent oxidation of CO to $CO_2$ is by reaction with $NO_x$ of OH as mentioned above. If the fuel is $CH_2O$ instead of $CH_4$, the latter stages of this chain become dominant. If the fuel is $C_2H_2$ instead of $CH_4$, then reactions of $CH_2$ become more important and $CH_2O$ becomes less important. Rate constants for the reactions involved in the hydrocarbon chemistry derived from previous studies are generally successful in the description of the transformation of $CH_4$ to CO.

The third major aspect of fuel/$NO_x$ flame chemistry is the interaction of H, C and N containing species. Reaction of $CH_1$ species with NO to form HCN and subsequent reaction of HCN, CN or NCO with NO lead to the conversion of NO to $N_2$. This process is obviously of most importance when $NO_2$ or NO is the oxidizer rather than when $N_2O$ is oxidizer, since in the latter case $N_2$ is formed directly. This scheme also shows the essential features of the oxidation process in the case where HCN is formed as a fuel during the decomposition of the energetic solid.

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