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TECHNICAL REPORT BRL-TR-2906

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GAS PHASE COMBUSTION CHEMISTRY OF  
NITRAMINE PROPELLANTS

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ANDRZEJ W. MIZIOLEK  
CARL F. MELIUS  
LARRY R. THORNE  
PAUL J. DAGDIGIAN  
MILLARD H. ALEXANDER

MARCH 1988

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## REPORT DOCUMENTATION PAGE

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| 19. ABSTRACT (Continue on reverse if necessary and identify by block number)<br>The details of the chemical processes which occur during propellant combustion are generally not known, yet, they directly affect such important propellant combustion characteristics as ignition and burn rate. Recently, a collaboration has been set up to study these details in a coordinated and comprehensive way. This study is concentrating on nitramine-based propellants since their ignition and burn rate properties are of great current interest and importance.<br><br>This report presents initial results of a long-term study in which both theoretical and experimental efforts are being pursued. Major results in theory include the calculation of thermochemical properties of a number of nitramine decomposition products and combustion intermediates, as well as specifying pathways for the formation of the reactive species HCN, H <sub>2</sub> CO, NO <sub>2</sub> , and N <sub>2</sub> O. Also, quantum chemical models for the interaction of nitrogen-type oxidizers with H atoms have been developed. On the experimental side, profiles of chemical |       |                                                  |                                                                                                              |                                       |                                  |
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19. Abstract (Cont'd):

species in CN-containing flames with  $\text{NO}_2$  as the oxidizer have been determined. Also, the dynamics of certain key propellant combustion reactions, which involve the H atoms as reactants, have been investigated. These results taken together represent a major advancement in our understanding of nitramine propellant combustion chemistry.

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## I. INTRODUCTION

The gun ballistic cycle is a complex phenomenon involving features of a number of technical disciplines including physics, chemistry, and mechanics, amongst others. Propellant combustion chemistry plays a very important role since it directly affects parameters such as ignition and the subsequent heat release which, in turn, affects properties such as muzzle velocity. A comprehensive understanding of propellant combustion chemistry has eluded the technical community for some time due to the extremely hostile environment for conducting chemical measurements. Currently, there is a resurgence of activity and expectations in this area due to a number of factors. Primary amongst these are the improvements and new developments in highly sophisticated diagnostic tools, such as lasers and mass spectrometers, as well as the recent involvement of highly capable laboratories, including the Sandia National Laboratory's Combustion Research Facility, Livermore, which is funded through the DA/DOE Memorandum of Understanding (MOU). The conditions, thus, exist for a concerted and comprehensive effort towards the understanding of propellant combustion chemistry.

This report presents early results of a multi-year research program which involves a collaboration between two universities and the DA and DOE laboratories. Due to the current development of low vulnerability propellants (LOVAs), research on nitramine propellants is especially important and timely particularly in regards to their ignition and burn rate characteristics. Both of these factors are directly related to the detailed chemical events involved in the initial activation and subsequent heat-releasing steps. Our research strategy is to study these detailed events through both theoretical and experimental means. Specifically, we report here the results of a comprehensive effort in theoretical thermochemical calculations, laboratory flame measurements, elementary reaction studies, as well as quantum chemical models. Together, these results represent a substantial coordinated program in the understanding of nitramine propellant combustion chemistry.

## II. RESULTS AND DISCUSSION

### A. Theoretical Thermochemistry Calculations

The energetics of chemical reactions and bond breaking require a knowledge of the thermochemistry of the various gas phase molecular species than can occur. In particular, the thermal stability of the short-lived, highly reactive radical species occurring during the combustion processes must be determined. The activation barriers of possible reaction pathways involving these combustion intermediates also must be determined. We, therefore, have applied the theoretical quantum chemical approach known as the BAC-MP4 method<sup>1,2</sup> to calculate the thermochemical properties of the molecular species. The approach involves the electronic structure calculation of a given molecule using the Hartree-Fock method. This technique is used to determine the optimum molecular geometry and harmonic oscillator vibrational frequencies. Total electronic energies are then calculated at a higher level of theory using Moller-Plesset many-body perturbation theory to fourth order (MP4). This method extends Hartree-Fock theory to include electron correlation, which is important in evaluating bond energies. We then include bond-additive corrections (BAC) to obtain the resulting heats of formation.



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Combining this information with the moments of inertia of the molecule and the vibrational frequencies provides the thermochemical entropies and free energies of the various combustion intermediates.

The resulting heats of formation and free energies using the BAC-MP4 method are given in Table 1 for the simplest nitramines,  $\text{H}_2\text{NNO}_2$  and  $\text{CH}_3\text{NHNO}_2$ , along with possible decomposition products and combustion intermediates. The free energies are given at various temperatures from 300 K up to 1500 K which encompasses the temperature range of the primary combustion zone in which the nitramine decomposes and initial combustion processes convert the  $\text{NO}_2$  to  $\text{NO}$ .

Table 1. Calculated Heats of Formation  $\Delta H_f^\circ$  and Free Energies of Formation  $\Delta G_f^\circ$  at Various Temperatures Using the BAC-MP4 Method for Various Molecular Species Involved in the Decomposition of Nitramines. (Energy in  $\text{kcal}\cdot\text{mol}^{-1}$ , Temperature in  $^\circ\text{K}$ )

| Molecular Species                          | $\Delta H_f^\circ$ |       | $\Delta G_f^\circ$ |       |       |       |
|--------------------------------------------|--------------------|-------|--------------------|-------|-------|-------|
|                                            | 0                  | 300   | 300                | 600   | 1000  | 1500  |
| $\text{H}_2\text{NNO}_2$                   | 5.8                | 2.4   | 20.6               | 39.3  | 64.6  | 95.7  |
| $\text{HNN}(\text{O})\text{OH}$            | 14.8               | 11.5  | 29.4               | 47.6  | 72.2  | 102.6 |
| $\text{H}_2\text{NONO}$                    | 23.5               | 20.5  | 37.6               | 55.1  | 78.7  | 102.6 |
| $\text{CH}_3\text{NHNO}_2$                 | 6.2                | 1.5   | 26.2               | 51.9  | 86.9  | 130.3 |
| $\text{CH}_3\text{NN}(\text{O})\text{OH}$  | 13.6               | 8.9   | 33.3               | 58.6  | 93.0  | 135.6 |
| $\text{CH}_3\text{NHONO}$                  | 26.7               | 22.4  | 45.9               | 70.1  | 103.0 | 143.7 |
| $\text{CH}_3\text{CH}_2\text{NO}_2$        | -19.2              | -24.2 | -2.5               | 20.0  | 51.1  | 90.0  |
| $\text{H}_2\text{NNO}$                     | 21.2               | 18.8  | 30.8               | 43.1  | 59.9  | 80.7  |
| $\text{CH}_3\text{NHNO}$                   | 21.6               | 17.6  | 36.2               | 55.8  | 82.6  | 115.8 |
| $\text{CH}_2\text{NNO}_2$                  | 36.6               | 33.6  | 50.0               | 66.5  | 88.7  | 116.2 |
| $\text{H}_2\text{NN}(\text{O})\text{OH}$   | 24.5               | 20.8  | 41.3               | 62.5  | 91.0  | 126.3 |
| $\text{CH}_3\text{NHN}(\text{O})\text{OH}$ | 25.2               | 20.0  | 47.4               | 75.7  | 114.1 | 161.6 |
| $\text{CH}_3\text{NHNHO}$                  | 40.4               | 35.5  | 57.9               | 81.3  | 113.6 | 153.7 |
| $\text{NH}_2\text{NHO}$                    | 37.9               | 34.6  | 50.3               | 66.6  | 89.0  | 116.6 |
| $\text{N}_2\text{O}$                       | 20.4               | 19.5  | 24.9               | 30.2  | 37.3  | 45.9  |
| $\text{CH}_2\text{O}$                      | -25.0              | -25.9 | -24.5              | -22.6 | -19.4 | -14.9 |
| $\text{H}_2\text{O}$                       | -57.1              | -57.8 | -54.7              | -51.2 | -46.2 | -39.5 |
| $\text{HCN}$                               | 32.4               | 32.2  | 29.8               | 27.4  | 24.4  | 20.8  |
| $\text{NO}_2$                              | 7.2                | 6.5   | 10.9               | 15.4  | 21.5  | 29.0  |
| $\text{HONO}$                              | -17.9              | -19.5 | -11.3              | -2.9  | 8.6   | 22.7  |
| $\text{NH}_2$                              | 46.8               | 46.1  | 48.3               | 50.9  | 54.6  | 59.5  |
| $\text{NO}$                                | 20.8               | 20.8  | 20.3               | 19.8  | 19.1  | 18.2  |
| $\text{H}$                                 | 51.6               | 52.1  | 48.5               | 44.8  | 39.5  | 32.5  |
| $\text{OH}$                                | 9.5                | 9.5   | 8.7                | 7.9   | 7.0   | 5.9   |
| $\text{HNO}$                               | 27.0               | 26.3  | 29.3               | 32.7  | 37.5  | 43.6  |
| $\text{CH}_3\text{NH}$                     | 47.7               | 45.0  | 52.7               | 61.2  | 73.8  | 90.0  |
| $\text{CH}_3$                              | 35.1               | 34.4  | 34.6               | 35.1  | 36.3  | 38.3  |
| $\text{H}_2\text{CN}$                      | 57.6               | 56.7  | 57.2               | 58.0  | 59.7  | 62.0  |
| $\text{CH}_3\text{NNO}_2$                  | 59.7               | 56.0  | 75.7               | 96.0  | 123.4 | 157.2 |
| $\text{HNNNO}_2$                           | 65.1               | 62.9  | 75.7               | 88.7  | 105.9 | 126.9 |
| $\text{CH}_3\text{NNO}$                    | 55.3               | 52.3  | 66.2               | 80.9  | 101.2 | 126.4 |
| $\text{HNNNO}$                             | 57.4               | 55.9  | 63.2               | 70.5  | 80.3  | 92.4  |

Using the BAC-MP4 method, one can also calculate transition state structures (saddle points on the potential energy surface) corresponding to bottlenecks in the reaction pathways. The resulting BAC-MP4 energies provide activation energies for reaction. The decomposition pathways for the reaction of H atom with  $\text{H}_2\text{NNO}_2$  are shown in Figure 1.

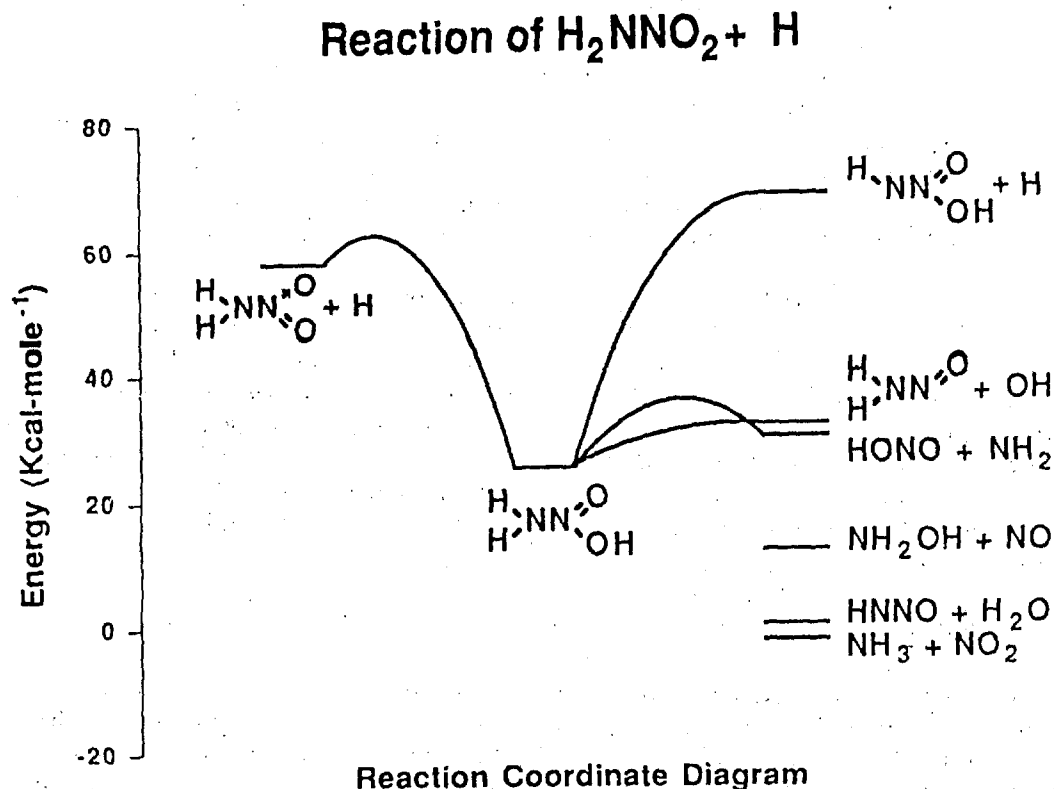
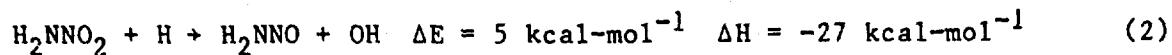
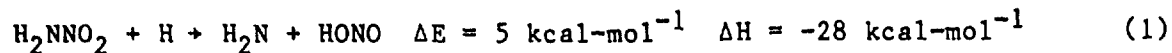


Figure 1. Calculated Reaction Pathways for the Reaction of  $\text{NH}_2\text{NO}_2 + \text{H} \rightarrow \text{Products}$ , Based on BAC-MP4 Heats of Formation at  $0^\circ\text{K}$  for Stable Species and Transition State Activated Complexes. Vertical energy scale is in  $\text{kcal-mol}^{-1}$ .

The results of these calculations indicate that the weakest bond in the nitramines is the  $\text{N-NO}_2$  bond, with a bond energy of  $48\text{--}49 \text{ kcal-mol}^{-1}$ . While there is considerable variation in the  $\text{N-N}$  bond energy proposed in the literature,<sup>3</sup> our value is in good agreement the estimates of Shaw and Walker.<sup>4</sup> Rearrangement of the molecule can also occur, leading to decomposition of the nitramine molecule. However, such elimination processes still have large activation energies of greater than  $40 \text{ kcal-mol}^{-1}$ . The HONO elimination from methyl-nitramine,  $\text{CH}_3\text{NHNO}_2$ , is calculated to be  $41 \text{ kcal-mol}^{-1}$ . Thus, the unimolecular decomposition of nitramines involves large energy barriers.

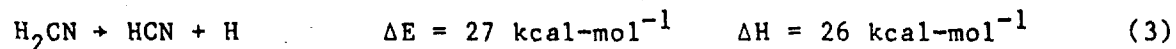
Meanwhile, the combustion process above the surface of the propellant is producing radical species as combustion intermediates. In particular, one such species is the hydrogen atom. In Figure 1, we consider the chemical

reaction of hydrogen atom with the nitramine,  $\text{H}_2\text{NNO}_2$ . One can see from the figure that the hydrogen atom can attack the oxygen of the nitro group, producing an energetically unstable intermediate which can decompose by scissioning either the N-N bond or the N-O bond, i.e.,



where  $\Delta E$  is the activation energy of reaction and  $\Delta H$  is the enthalpy of reaction. In both cases, the reaction is exothermic (28 and 27  $\text{kcal-mol}^{-1}$  respectively) and the required activation energy is small (5  $\text{kcal-mol}^{-1}$ ), corresponding to the energy barrier for H atom attacking the nitro group. Thus, we see that the H atoms provide an alternative, low activation energy reaction pathway for the decomposition of nitramines.<sup>5</sup>

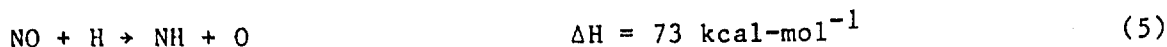
Having broken the N-N bond (either by unimolecular decomposition from the heat generated in the primary flame zone or from reaction with H atoms generated in the primary flame zone), the remaining bond energies are significantly reduced. In particular, the C-N bond energy is reduced from 80-85  $\text{kcal-mol}^{-1}$  to less than 20  $\text{kcal-mol}^{-1}$ . Thus, once the  $\text{NO}_2$  group has been removed, the HMX or RDX molecule will continue to fall apart to form  $\text{H}_2\text{CNNO}_2$ 's and  $\text{H}_2\text{CN}$ . The  $\text{H}_2\text{CNNO}_2$  molecule, whose N-N bond energy is 28  $\text{kcal-mol}^{-1}$ , will break apart to form more  $\text{H}_2\text{CN}$  and  $\text{NO}_2$ . The  $\text{H}_2\text{CN}$  will decompose in the primary flame zone to form  $\text{HCN} + \text{H}$ , i.e.,



Thus, we see that chemical reaction processes occurring in the primary flame zone of the nitramine combustion can produce a source of H radicals which can aid in the initial decomposition step of the combustion process. Since the concentration of H atoms consumed in Reactions (1) and (2) are regenerated by Reaction (3), the H atoms serve as an autocatalytic species in the combustion process aiding the decomposition.<sup>3</sup> Consistent with this mechanism, the calculations support experimental data<sup>3</sup> that indicate that  $\text{HCN}$  and  $\text{NO}_2$  should be important gas phase combustion species in burning of nitramines.

Meanwhile, Reaction (2) leads to nitrosamines, which have been observed as intermediate products in HMX and RDX decomposition,<sup>3</sup> and to OH radicals. The nitrosamines can lead to  $\text{N}_2\text{O}$  formation, while OH can react with the nitramines and intermediate products through abstraction and addition reaction pathways to form  $\text{H}_2\text{CO}$ . Thus,  $\text{H}_2\text{CO}$  and  $\text{N}_2\text{O}$  could also be important gas phase combustion species.

The theoretical BAC-MP4 thermochemical calculations have indicated an important role that H atoms can play in the combustion of nitramines. However, the combustion process is very complicated, with many other radical species being generated in the flame zones. Also, the H atoms and other radicals can react further with the combustion intermediates. For instance, H atoms react very rapidly with  $\text{NO}_2$  to form  $\text{NO} + \text{OH}$ . The H atom can react with the NO, producing the secondary flame. However, this reaction is very endothermic,



and competing reactions can inhibit this secondary flame. Thus, one needs to know the various reaction mechanisms which can occur in the flame and the resulting combustion intermediate species concentration in the flame. The experimental and theoretical techniques discussed next provide needed information regarding these processes.

## B. Laboratory Flame Measurements

We have initiated studies of a series of low pressure flames related to the combustion of the gas phase species above burning nitramines. These species can be broadly classified as fuels or oxidizers. The fuels include HCN and  $\text{H}_2\text{CO}$ , and the oxidizers include NO,  $\text{NO}_2$ , and  $\text{N}_2\text{O}$ . Our initial studies are centered on the combustion of HCN and  $\text{NO}_2$ . This choice is motivated by the theoretical results that show these species could be important products of the high temperature pyrolysis of the solid nitramines, as well as by previous experimental work.<sup>3,5</sup>

Our studies use the same techniques and methods successfully employed previously to study low pressure Ar- $\text{H}_2$ - $\text{O}_2$  flames doped with various combinations of  $\text{C}_2\text{H}_2$ , HCN, and NO.<sup>6,7</sup> These studies are made on flames operated at low pressure near the blow-off limit in order to minimize the heat transferred from the flame to its surroundings. This situation allows model calculations to be more easily compared to the experimental data. The primary objective of these studies is to determine the chemical mechanism of the flames under investigation; that is, to determine which chemical reactions are the most important in the process of converting reactants into products. Because of the large number of species and reactions involved in the combustion process, the combustion mechanism is elucidated by making detailed flame model calculation that are then compared to the measured temperature and species profiles. Reaction rate constants are obtained from the literature or are estimated with the aid of theoretical thermochemical calculations if literature values are not available. These estimates are revised, within reasonable limits, until the results of the model calculations agree with the observed data.

The experimental apparatus includes: (1) a premixed, flat flame burner having a 9.7-cm diameter porous surface and operated at 25-Torr ambient pressure, (2) a microprobe sampling mass spectrometer to measure the concentration of abundant stable species, (3) a laser induced fluorescence (LIF) spectrometer to measure the concentrations of CN and CH radicals and to measure the CN rotational temperature, and (4) a Fourier transform infrared spectrometer (FTIR) to measure concentrations of stable species and abundant free radicals. The burner is translated vertically in the vacuum housing in order to obtain temperature and species profiles. The position of the burner relative to the analytical probes is known to within 0.02 cm.

The mass spectrometer system is mounted on top of the burner vacuum housing to minimize the distance between the burner and the spectrometer. The sampling system consists of a 1-cm quartz probe drawn to a tip having a 0.01-cm diameter opening. This probe is pumped by an auxiliary vacuum pump

that maintains the probe pressure near 1.5 Torr. The combustion gases entering the probe are directed through a microchannel plate, a chopper, a skimmer and into the ionization region of a quadrupole mass spectrometer. The ion signal is then detected with a lock-in amplifier. This discriminates against gases trapped in the mass spectrometer vacuum housing. The mass spectrometer sampling system has a lower sensitivity limit of 10-50 ppm with a typical relative accuracy of 1% for concentrations about 1000 ppm. The sensitivity of the mass spectrometer is calibrated for each species to be measured immediately before acquiring the flame profile.

The LIF spectrometer is composed of a modified dye laser pumped by a cw, argon ion laser. The dye laser uses polyphenyl-1 dye and operates in the 350-400-nm region. CN concentration profiles are measured by scanning over the B-X,  $R_1(14)$  and  $R_2(14)$  transitions twice and averaging their intensities. The same procedure is used for measuring relative CH concentrations except that the B-X,  $Q_1(5)$  and  $Q_2(5)$  transitions are used. The flame temperature profile is obtained from the rotational temperature of the CN which is determined from the  $R_1$ ,  $R_2$  rovibrational band contour. The FTIR is a commercial instrument and operates in the 400-4000- $\text{cm}^{-1}$  region. The maximum resolution is 0.004  $\text{cm}^{-1}$ .

Typical data obtained with the mass spectrometer system is shown in Figure 2 which shows stable species profiles for a  $\text{C}_2\text{N}_2$ - $\text{NO}_2$  flame. This flame represents a subset of nitramine combustion chemistry, addressing those reactions involving primarily CN radicals in the absence of hydrogen. The data show that NO is produced as an intermediate (reaching a peak value at 1.5 cm) and that  $\text{C}_2\text{N}_2$  is not depleted to any great extent until the  $\text{NO}_2$  is consumed. We are presently developing a chemical kinetic model that will aid in interpreting these data and that will help to identify the most important reactions occurring in the flame. However, even without the model it appears that NO produced from  $\text{NO}_2$  plays a major role in the oxidation of  $\text{C}_2\text{N}_2$ .

The  $\text{C}_2\text{N}_2$ - $\text{NO}_2$  flame has several unusual characteristics. It produces bright visible emission in three distinctly colored bands: yellow (0.7-0.9 cm), blue (0.9-1.1 cm), and pink (1.1-2.4 cm). The spatial relationship of these bands are indicated in Figure 3 which is a black and white photograph of the flame. Premixed flames generally do not show this banded structure because the fuel and oxidizer are thoroughly mixed prior to combustion, and thus, there are no large concentration gradients to produce a banded flame structure. An additional unusual aspect of this flame is that there are no hydrogen-containing free radicals to support the combustion process as there are in most other flames.

### C. Elementary Reaction Studies

In order to gain some insight into the reaction of hydrogen atoms with propellants, we have investigated three prototype elementary reactions under low pressure single-collision conditions:



## CYANOGEN - NITROGEN DIOXIDE FLAME

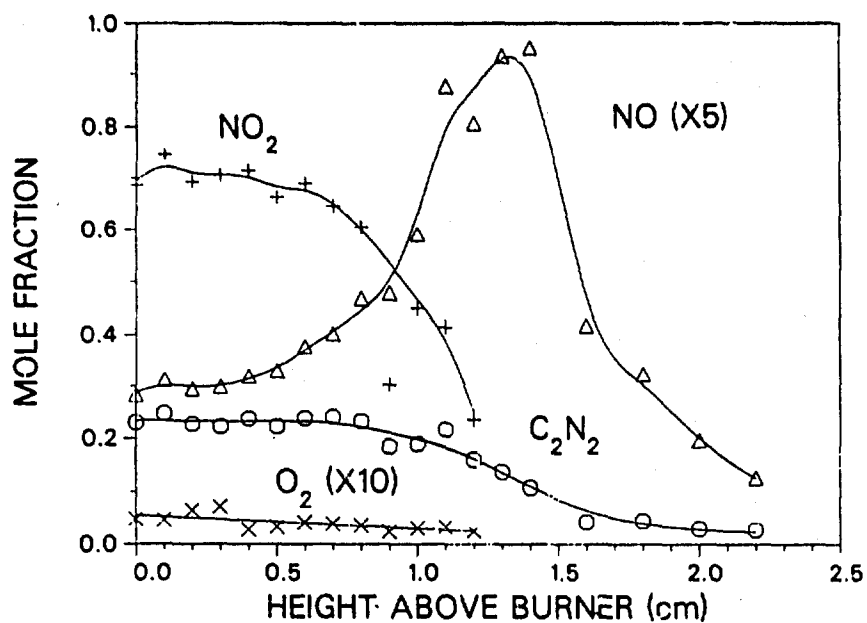


Figure 2. Stable Species Profiles of a Low Pressure  $C_2N_2$ - $NO_2$  Flame

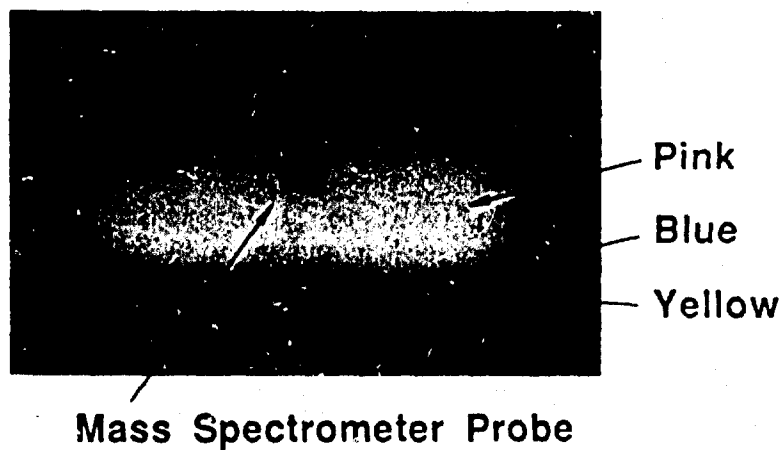


Figure 3. Schematic Diagram of a Low Pressure  $C_2N_2$ - $NO_2$  Flame. Colors of the three emitting regions are indicated.

The nascent OH product internal energy distribution has been measured for Reaction (6) in several laboratories by uv laser fluorescence<sup>8,9</sup> and infrared chemiluminescence<sup>10,11</sup> detection. The product translational energy distribution has also been measured in crossed beam scattering experiments.<sup>12,13</sup> The OH internal energy and the product recoil energy account for about 68% of the total energy, the remainder presumably residing in internal excitation of the NO product. In view of the importance of this reaction, we have undertaken to measure its internal energy distribution by uv laser fluorescence.

An important feature of Reactions (7) and (8) is that there are a number of different exothermic reaction pathways available. We have studied these reactions by laser fluorescence detection in order to estimate product internal energy distributions and branching ratios, which are extremely uncertain. It should be noted that, while this detection technique is very convenient and sensitive, it is not universally applicable. We apply it here to the detection of OH, CH<sub>3</sub>O, and NO.

These studies were carried out in a beam-static gas scattering arrangement at total pressures of 2 mTorr or less. A beam of atomic hydrogen, prepared by dissociation of H<sub>2</sub> in a microwave cavity, passes through an orifice into a reaction chamber to which the reaction partner is added. Reaction products were monitored by laser fluorescence excitation with the frequency doubled and/or mixed output of a Nd:YAG pumped dye laser.

The apparatus was first tested by observing the OH product in the ground  $v=0$  vibrational level and comparing with the previously reported results.<sup>8,9</sup> In agreement with the literature, we find nascent OH products peaked near the rotational quantum number  $N=14$ , but most of the OH detected was collisionally relaxed to low  $N$  levels, presumably because of collisions with the walls. Relaxation effects are much less important for vibrationally excited products since wall collisions efficiently quench vibrational energy.

We were also able to detect OH product from the  $H + CH_3NO_2$  reaction. Because of the small rate constant for this reaction,<sup>14</sup> the fluorescence signal was considerably smaller than for Reaction (6). The detection of an albeit weak OH signal from this relatively slow reaction nevertheless implies that the formation of OH is a significant reactive pathway. This is consistent with the expectation that the hydrogen will interact most strongly with a lone pair of an oxygen atom of the CH<sub>3</sub>NO<sub>2</sub> reagent.

It is interesting to note that only low  $N$  OH rotational levels are populated in Reaction (7), and moreover only the lower  $^2\Pi_{3/2}$  spin component is present. It is unlikely that the presence of only one spin component is an artifact of possible collisional relaxation, but rather must reflect the nascent product distribution. The observed small degree of product rotational excitation for this reaction is similar to that observed previously in the reaction of oxygen atoms with a number of hydrocarbons.<sup>15,16</sup> This small degree of rotational excitation has been interpreted as showing that reaction occurs when the oxygen atom approaches collinear to a C-H bond. In these reactions, as in Reaction (7), a significant preference for formation of the lower OH spin component was observed. This can be explained by assuming that the reaction proceeds adiabatically along the lowest-energy potential surface, which correlates with OH  $^2\Pi_{3/2}$  products.

Reaction (8) was also studied, but no laser fluorescence signals due to reaction products (OH, CH<sub>3</sub>O) could be detected. However, a weak unstructured fluorescence signal could be observed with just the methyl nitrite reagent present. On the basis of an earlier study of the uv photolysis of methyl nitrite,<sup>17</sup> this signal is assigned as a two-step process involving CH<sub>3</sub>ONO photolysis and subsequent fluorescence excitation of CH<sub>3</sub>O fragment. This background signal could be obscuring the observation of reaction products.

The NO product in the  $v=0$  vibrational level from the H + NO<sub>2</sub> reaction was also observed. Figure 4 shows a fluorescence excitation spectrum of the A-X (0,0) band near 226 nm. Comparison of this spectrum with that for a room temperature NO sample indicates that the product state distribution is remarkably close to a 300 K Boltzmann distribution or, more likely, that there is considerable collisional relaxation of the NO product. However, from comparison of the relative intensities of the P<sub>1</sub> and Q<sub>1</sub> vs. P<sub>2</sub> and O<sub>12</sub> band heads we find that the  $^2\Pi_{1/2}$  and  $^2\Pi_{3/2}$  spin components are equally populated in the reaction. This lack of spin equilibrium is consistent with the known order of magnitude difference in spin-changing vs. spin-conserving inelastic collisions involving NO.<sup>19,20</sup>

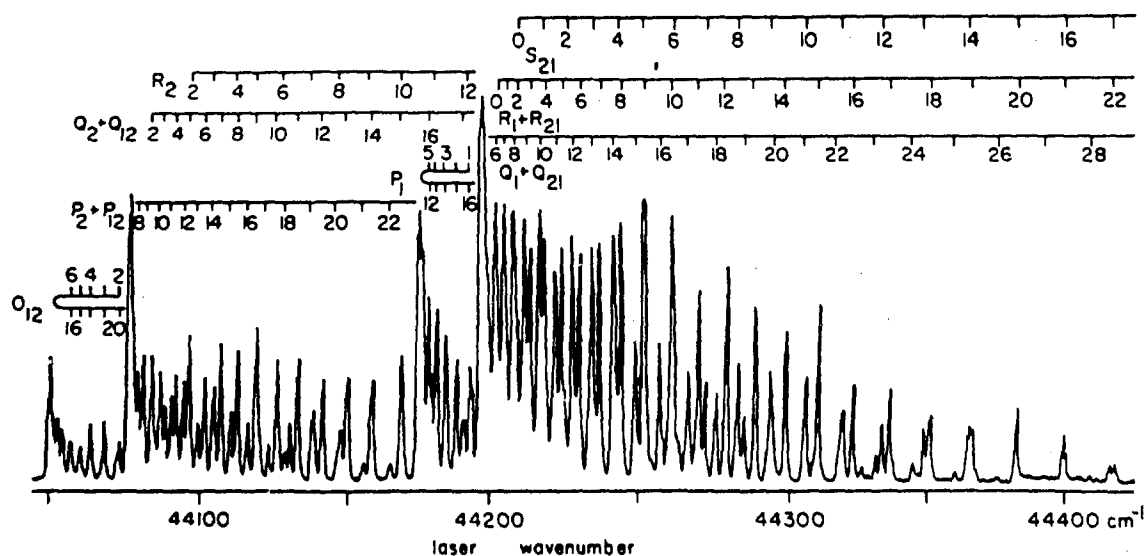


Figure 4. Excitation Spectrum of NO Formed in the H + NO<sub>2</sub> Reaction. The NO<sub>2</sub> gas pressure was 0.15 mTorr. Lines of the NO A-X (0,0) band are identified.

The observation of equal population of the spin-orbit levels of both OH ( $X^2\Pi$ ) previously<sup>8,9</sup> and NO ( $X^2\Pi$ ) in the present work implies that there is no preferential orientation of the spin angular momentum with respect to the plane of rotation. While Reaction (6) is believed to proceed through an intermediate HONO complex on the lowest  $^1A'$  surface, nonadiabatic processes will mix this surface with the other exit channel surfaces (8 in all) as their energies asymptotically approach one another. The presence of a significant amount of product rotational excitation suggests that this mixing will be

efficient, leading to a statistical distribution of spin states in both reaction products.

#### D. Quantum Chemical Models

Collaborative theoretical work has concentrated on inelastic  $H + NO$  collisions, which have been studied experimentally by Leone and coworkers.<sup>21</sup> This process may well play an important role in the observed rapid relaxation of the NO product in the  $H + RNO_2$  reactions reported here. Recently, Colton and Schatz<sup>22</sup> have carried out quasiclassical trajectory studies of collisions of H with NO ( $X^2\Pi$ ) at translational energies of 1-3 eV, using semi-empirical fits to the four potential surfaces ( $^1A'$ ,  $^3A'$ ,  $^1A''$ ,  $^3A''$ ) which correlate with the lowest energy  $H + NO$  asymptote. Unfortunately, a classical trajectory study can give no information either on the aspects of the collision dynamics which reflect the open-shell nature of the collision partners, namely to what extent will energy transfer depend on the fine-structure level<sup>23</sup> of the NO, or on quantum interference between the four surfaces.

Collision-induced transitions between the multiplet and/or rotational levels of NO scattered by an atomic target arise because the orbital motion of the collision partners couples with the nuclear rotational, electronic orbital, or electronic spin angular momenta of the molecule. An additional degree of complexity is introduced when the target is also an open-shell species, because potential surfaces of more than one spin multiplicity will contribute to the collision dynamics.<sup>24,25</sup> A particularly intriguing question is the extent to which the correlation of the nonzero spin of the target with the spin of the molecule will result in a breakdown of the propensity rules which we have derived theoretically<sup>26</sup> and which have been observed experimentally<sup>27,28</sup> for collisions of molecules in  $^2\Pi$  electronic states with closed-shell atoms.

We have begun the development of the quantum description of the collision of a  $^2\Pi$  molecule with a  $^2S$  atom. A first key step involves projection of the adiabatic electrostatic interaction potential onto the diabatic molecular and atomic electronic spin subspaces by use of a tensor expansion.<sup>24</sup> It is also necessary to make use of the recoupling scheme of Corey and McCourt,<sup>29</sup> and to treat the rotor molecular rotational wavefunctions in Hund's case (b) rather than case (a).<sup>23</sup>

Even at moderate energies the motion of the light H atom will be rapid enough to justify use of the infinite-order-sudden (IOS) approximation.<sup>26,30-32</sup> In this limit the rotational motion of the NO molecule is considered to be effectively frozen during the collision. The collision cross sections can then be written as integrals of a sudden transition operator over the H-NO orientation angle. The electrostatic potentials of Colton and Schatz<sup>22</sup> can be used directly, without re-expansion in a series involving Legendre polynomials as would be necessary in a full close-coupled treatment of the collision.<sup>26</sup> Application of the recoupling scheme<sup>29</sup> to NO ( $X^2\Pi$ ) will necessitate ignoring the energy splitting between the two spin-orbit manifolds, which certainly is entirely consistent with the IOS approximation. We have already begun numerical calculations to investigate the validity of this approximation for the simpler case of collisions of NO ( $X^2\Pi$ ) with Ar.

### III. CONCLUSION

The multi-laboratory collaboration on nitramine propellant combustion chemistry is yielding significant new results in both the theoretical and experimental areas. On the theoretical side, great progress has been made in the calculation of thermochemical properties of nitramine decomposition products and combustion intermediates using ab initio quantum chemical methods. This work supports the evidence that HCN and NO<sub>2</sub> play a major role in propellant flames and specifies pathways for the formation of other important reactive species such as H<sub>2</sub>CO and N<sub>2</sub>O. In addition to the thermochemical calculations, a quantum chemical framework has been set up to describe the details of collisional interactions between NO molecules and H atoms.

On the experimental side, the profiles of chemical species in the C<sub>2</sub>N<sub>2</sub>/NO<sub>2</sub> flame have indicated the importance of NO in the oxidation of the fuel molecules. Also, detailed elementary reaction studies have yielded important information concerning the dynamics of key propellant combustion reactions involving the H atom and NO<sub>2</sub>-containing oxidizers. In summary, these results represent major progress in the understanding of propellant combustion chemistry.

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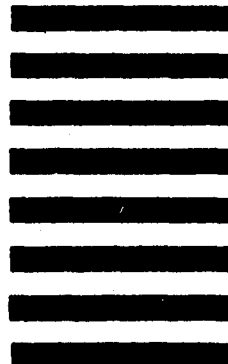


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