Lawrence Berkeley National Laboratory
Recent Work

Title
PULSED PLASMA JET IGNITERS: MOLECULAR BEAM MEASUREMENTS

Permalink
https://escholarship.org/uc/item/33v3w2n3

Author
Cavolowsky, J.A.

Publication Date
1985-10-01
Presented at the Western States Section/
The Combustion Institute Conference,
Davis, CA, October 21-22, 1985

PULSED PLASMA JET IGNITERS:
MOLECULAR BEAM MEASUREMENTS

J.A. Cavolowsky, C.F. Edwards,
A.K. Oppenheim, and D. Lucas

October 1985
DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.
Pulsed Plasma Jet Igniters: Molecular Beam Measurements

J.A. Cavolowsky, C.F. Edwards*, A.K. Oppenheim and D.Lucas

Applied Science Division
Lawrence Berkeley Laboratory
University of California
Berkeley, CA 94720

*present address
Combustion Applications Division
Sandia National Laboratory
Livermore, CA 94550

Meeting of the Western States Section of the Combustion Institute
October 1985

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Engineering, Mathematics and Geosciences Division, and the Assistant Secretary for Conservation and Renewable Energy, Office of Transportation Programs, Division of Transportation Energy of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.
ABSTRACT

Molecular beam mass spectrometry has been used to determine temperature and species concentrations in the discharge plume of a pulsed plasma jet igniter. Laser schlieren photographs of the plume/sampling cone interaction reveal that the molecular beam sampling has negligible effect on jet fluid mechanics up to the time of impingement, and temperature and species measurements indicate that at later times the gas is sampled from within the plume. The temperature in an atmospheric pressure jet of nitrogen ranged from a maximum of 50,000K to ambient temperature. Peak nitrogen atom concentration at the detector was found to range from 5%, 2mm from the igniter, to less than 100 ppm at 14mm.
INTRODUCTION

Pulsed plasma jet igniters produce a transient jet capable of initiating and enhancing combustion (Oppenheim et al., 1978, Boston et al., 1984, Edwards et al., 1983). The success of the plasma jet igniter has been attributed to its ability to move the source of ignition from proximity to cold surfaces, to entrain ambient combustible gases (increasing the mass of the ignition kernel), and to spread reactive species throughout the combustible gas. Experimental and analytical studies have characterized the penetration and mass entrainment (Cetegen et al., 1980, Topham et al., 1982). However, radical species seeding is difficult to quantify due to problems inherent in the detection of species in the transient, inhomogeneous flow field of the pulsed plasma jet.

Topham et al. (1975) determined the temperature in the luminous portion of an atmospheric pressure air plasma jet by measuring the ionization current collected by a probe inserted into the plume. By assuming thermal and chemical equilibrium, and measuring the plasma streaming velocity, the temperature was found. Unfortunately, this method is only sensitive in the region near the orifice of the jet where the ion density is high. As such, these measurements were restricted to the first 15mm of the plasma plume, where the temperature was found to be in the range from 9500 to 2500K.

Clements et al. (1984) measured the temperature downstream
of the luminous zone by means of a high spatial resolution interferometer. These measurements provided information about the temperature late in the development of the plume, after significant entrainment had occurred. Temperatures measured were in the range from 700 to 350 K.

There are no quantitative measurements of radical species concentration currently in the literature. However, deductive techniques have been used by Clements et al. (1984) to obtain information about the radicals present. A nitrogen plasma discharged into oxygen produced nitric oxide which was presumed to be formed by the reaction of nitrogen atoms and diatomic oxygen. The location of a recombination surface was varied and the effect monitored by the change in NO concentration. Using this technique, detectable levels of chemical activity were found as far as 30mm from the igniter.

Molecular beam mass spectrometry (MBMS) provides a means for quantitative examination of the species within the plume of the plasma jet igniter. Previous studies of both steady-state and transient systems have shown that MBMS allows for measurement of stable and unstable (radical) species and determination of temperature by the time-of-flight (TOF) method (Peterson et al., 1984, Lucas et al., 1984, Lucas et al., 1985, Sloane and Ratcliffe, 1984).

In this paper we present the application of MBMS and TOF techniques to the transient, inhomogeneous flow field of the
pulsed plasma jet igniter. Results are reported for the temperature and radical species concentration within the plume of a nitrogen pulsed plasma jet.

EXPERIMENT

The pulsed plasma jet igniter used in this study, shown schematically in Fig. 1, is the same one used in the engine studies of Edwards et al. (1985). An arc discharge is formed in the igniter cavity between the cathode (center electrode) and anode (discharge orifice). The current pulse is a slightly underdamped half-wave produced by an RLC circuit and has a duration of 100us. Total stored energy is 3J. Cavity diameter, length, and orifice diameter were set at 3mm.

The major components of the molecular beam apparatus have been described in detail previously (Lucas et al., 1984). The sampling probe is a 120 degree quartz cone, 4mm high with a 100um diameter orifice at its vertex. One significant change from the previous configuration involved moving the quadrupole assembly from a crossed-beam to an axial location. This increased the signal/background ratio by an order of magnitude and reduced the transit time through the assembly to 10us.

High speed laser schlieren photographs were taken with an apparatus described previously (Oppenheim et al., 1966). A Q-switched ruby laser was used to provide light pulses 50ns in
duration at a frequency of 200kHz. Images were recorded using a rotating mirror drum camera.

RESULTS AND DISCUSSION

1. Sampling Cone/Igniter Interactions

A major concern at the outset of this research was that the sampling system might significantly perturb the flow field of the plasma jet. While the sampling system had been successfully used previously to monitor homogeneous combustion (Lucas et al., 1985), it was not known a priori how the jet would interact with the sampling cone and the flow of gas through the orifice. Previous studies of direct sampling perturbations were focused on steady-state flame systems (Biordi et al., 1974, Yoon and Knuth, 1980), and not the transient, highly turbulent, inhomogeneous flow from the plasma jet igniter.

Figures 2 and 3 show a series of laser schlieren photographs of the plume/sampling cone interaction at various igniter/sampling cone distances. Each series of photographs taken at a fixed separation distance was recorded from a single igniter discharge. Figure 2 shows records from the first 30 microseconds after triggering the igniter, while Fig. 3 shows the later stages of jet development up to 235 microseconds. It should be noted that refractive index gradients are observed in
the schlieren photographs for several milliseconds. It appears that in the early stages of plume formation the presence of the cone has negligible effect, and that the plume front (the most advanced portion of the schlieren image) reaches the sampling cone in the same time as would be required for penetration to that depth without the presence of the sampling cone. At later times, when the front has advanced well past the sampling cone, significant differences are observed for the portion of the plume that has passed the cone, but the gas actually sampled does not appear to be disturbed by the deflection of the plume. However, this comment must be tempered since strong refractive index gradients in the outer regions of the plume (away from the sampling cone) may mask any disturbances localized at the cone. Additional photographic records (not shown) with no flow through the sampling orifice indicate that the presence or absence of sample flow has little effect on the plume structure.

To quantify the interaction, we measured the displacement of the blast wave and plume front for several igniter/sampling cone distances. These results are shown in Fig. 4. Two measurements of the plume penetration are presented; the maximum plume penetration and the maximum penetration on the igniter/sampling cone axis. Since the plume structure is asymmetric and stochastic, fluctuations exist in the axial displacement/time characteristic of the jet. However, as apparent in Fig. 4, the maximum penetration of the jet is quite repeatable and not significantly effected by the presence of the sampling cone.
2. Time-of-Flight Measurements

The time-of-flight technique has been used previously to determine temperature in molecular beam systems (Young et al., 1970, Peterson et al., 1984). However, the transient and inhomogeneous flow field of the plasma jet makes application of TOF techniques a difficult task. As such, we first expand on the TOF method and then present experimental results for nitrogen plasmas.

Before discussing the behavior of the sample within the molecular beam, it is necessary to develop an expression for the velocity which a packet of gas, sampled at the orifice, will achieve after expansion. Assuming that the flow downstream of the sampling orifice is well modeled as an adiabatic expansion of an ideal gas in translational equilibrium, the mean velocity of the sampled gas \( U \) is given by

\[
U = \sqrt{\frac{RT_0}{M}} \left( \frac{1}{M} + \frac{\gamma - 1}{2} \right)^{\frac{1}{2}}
\]

(1)

where \( T_0 \) is the stagnation temperature and \( M \) is the Mach number. For large Mach numbers this expression simplifies to

\[
U = \frac{2\gamma RT_0}{\gamma - 1} = \sqrt{2C_p T_0}
\]

(2)

This approximation introduces an error of less than 10% for \( \gamma = 1.4 \) and Mach numbers in excess of 5, conditions that are met in our apparatus for temperatures in the range of 300 to at least
3000 K (Lucas et al., 1985).

A displacement vs. time diagram of the molecular beam sampling process is used to illustrate the TOF process, and is shown in Fig. 5. The upper part of this figure illustrates flow through the system in the absence of beam chopping, while the bottom part introduces the TOF chopper into the system.

In the unchopped case gas is continuously sampled. The trajectories of the sampled gas after expansion are shown by straight lines with slopes equal to the inverse of velocity. Spacing of the trajectories in time is chosen to represent equal sampling intervals. Before the jet plume reaches the sampling cone only ambient gas enters the system. The trajectories are parallel (denoting constant temperature and therefore velocity) and travel the length of the beam path in a time given by $X_{od}/U(T_a)$. Once the sample reaches the ionizer assembly it is ionized and accelerated to a high velocity (accelerating potential typically 10-15 eV) before traveling through the quadrupole to the detector. The acceleration of the ions assures that the flight time through the ionizer/quadrupole/detector assembly is essentially constant regardless of the initial velocity (temperature) of the sample. The trajectories through this region thus have the same slope, indicative of the post-ionizer velocity. The flux of molecules reaching the detector is given by the density of lines at the right hand side of the figure, with a uniform spacing indicative of a steady-state signal.
Some time after the beginning of the igniter discharge, the jet will impinge on the sampling cone and enter the molecular beam. Assuming that the gas in the plasma jet is at a higher temperature than ambient, the velocity of this sample will be higher according to Eq. 2. The hot gas may actually pass the lower velocity ambient gas during transit down the beam path, resulting in its arrival at the detector first (beam overrun) (Peterson et al., 1984). As sampling of the jet proceeds, the hotter gas from the jet will continue to overrun the cool ambient gas until the last portion of ambient gas has completed its travel to the detector. At this point the sample arriving at the detector is due only to the plasma jet with no contribution from the ambient gas; as a result the signal at the detector will decrease. In addition, if the temperature of the gas at the source decreases, the detected signal will drop below the original baseline level, as evidenced by the spreading of the gas packet trajectories.

The signal expected for a species present in both the ambient medium and in the plume is shown at the far right of the figure where both over- and underrun effects are evident. If the species is present only in the plume (e.g., a radical species) over- and underrun will still occur in the beam but this will not be evident in the detector signal since the species concentration is zero before and after the plume molecules arrive at the detector. Nevertheless, this must be taken into account when calculating the concentration of any species from the detector
Experimental data illustrating the above case is shown in Fig. 6. The upper trace is the molecular nitrogen signal (mass 28), while the lower trace is for atomic nitrogen (mass 14). The igniter, located 2 mm from the sampling cone, is discharged in nitrogen at atmospheric pressure. The chopper begins to open at 150 microseconds and remains open through the remainder of the event. The $N_2$ signal rises to a nearly constant level before the igniter is fired at 550 microseconds. Beam overrun is observed until the 1 ms mark, after which underrun begins to occur. Variations in the signal level continue for several milliseconds. In contrast, the $N$ atom signal, present only in the hotter portions of the plume, increases from its initial level (zero), peaks, and returns to zero well before the $N_2$ signal is steady.

Introducing a chopper with a narrow slit changes the TOF picture to that shown in the bottom half of Fig. 5. The situation depicted is one where a large temperature increase occurs at jet arrival. While the chopper permits molecules to pass only during a short time interval, the temperature (velocity) of the sample packets admitted by the chopper may cover a wide range. The appropriate detector signal for this case is shown at the right of the figure. The first peak observed is that from the hot plume while the second is from the colder ambient gas. The time required for the hot and cold packets to travel from the chopper to the ionizer is given by $X_{cd}/U(T)$ and $X_{cd}/U(T_a)$, respectively, where $X_{cd}$ is the chopper to
ionizer distance. Since both packets travel an identical distance, the ratio of flight times gives the velocity ratio of the packets and thus their temperature ratio. Determining the temperature of the hot packet requires only a measurement of the time interval between arrival of the packets at the detector (\( \Delta T \)), and the time required for the ambient gas to travel from the chopper to the ionizer, resulting in the expression:

\[
T = T_a/(1 - \frac{\Delta T \sqrt{2C_T} T_0}{x_{cd}})
\]

(3)

Experimental chopped beam \( N_2 \) signals are shown in Fig. 7. The triangle below each trace denotes the time the igniter fired. The chopper opens at the same time for each trace, as evidenced by the first two ambient temperature peaks in each run. Traces are ordered with the igniter firing 50 microseconds later for each curve, with the lowest trace showing sampling of the plume at the earliest time.

Temperatures in the plume were determined initially by using the peak arrival time method described above. After determining a single temperature for a particular peak in the TOF data, the time that this gas was sampled was calculated. The difference between this time and the igniter firing time is the plume propagation time. The results in Fig. 4 were then used to determine where in the plume the sample originated. These measurements are presented in Fig. 8, with each point obtained from a single experiment. The width of the chopper opening is
shown at the top of the figure, and is the same as the maximum uncertainty in locating the position of the plume at the time the data was recorded. The uncertainty in temperature varies due to the square root dependence of velocity on temperature, and is estimated to be 30K at 500K, 200K at 2000K and approximately 15,000K at 50,000K.

The maximum temperature was 50,000K 2mm from the igniter, 2900K at 4mm and 1670K at 8mm. The 50,000K point is not unreasonable in light of the electron and ion temperatures measured in arc discharges (Hellund, 1961). These high temperatures are also observed for atomic nitrogen, and for argon in 2% Ar/N2 mixtures. This hot gas would be rapidly cooled at the plume front, where the collision rate with colder molecules is very high. This would account for the steep drop of the maximum temperature past the 2mm point. Temperatures at distances greater than 4mm may also be higher than those measured, but signal/noise considerations may have prevented their observation. Behind the front, temperatures are much lower, and there is evidence for near ambient pockets entrained in the plume. This is not surprising as much evidence exists showing the turbulent nature of the plume.

In reality the analysis is not as simple as the above description suggests. In particular, the effects of large temperature gradients at the source, finite chopper opening time and the thermal velocity distribution of the gas in the beam complicate the procedure in most cases. In situations where
these factors must be taken into account, the detailed TOF simulation procedure described previously (Peterson et al., 1983, Peterson, 1984) may be used.

Results obtained by the peak arrival time method at 2mm igniter/sampling cone separation were used as a guide to produce the temperature profile shown in Fig. 10. This profile was used in the detailed TOF simulation to calculate the expected TOF curve for the different igniter firing times shown in Fig. 9. A comparison of the experimental (left) and calculated (right) results are shown in Fig. 11. Each curve represents a 50 microsecond change in the firing time of the igniter. It should be noted that our model does not include the effect of sample flowrate changes at the orifice with temperature, so the height of the curves will differ. However, the timing and general shape of the curves are in good agreement, and support our temperature determinations.

3. Species Measurements

The concentration of N atoms in the plume was determined as a function of time and distance from the igniter using continuous sampling. Electron energies of 20.5 eV were used for the atoms and 21.5 eV for molecular nitrogen. No measurable cracking of N$_2$ at this energy was observed at ambient conditions. The atom signals were averaged, normalized to averaged N$_2$ signals, and converted to mole fractions using the ionization cross-sections of Kiefer and Dunn (1966).
Results for the radical concentrations are shown in Fig. 11. The maximum concentration, 5%, is found at the shortest separation distance of 2mm. The peak value measured drops rapidly and smoothly with increasing distance, with no radicals measured above the detection limit (estimated to be 100ppm) for distances greater than 14mm. The time of peak concentration at a given distance also increases with separation distance as expected.

The concentrations measured for the atomic species are in reasonable agreement with the values expected for the equilibrated gas (Edwards, 1985). It must be remembered that the concentrations presented are in the detector time frame, and that the values are an average over many experiments and probably over a range of temperatures, preventing direct comparison with equilibrium calculations.

No attempt was made to measure the concentration of metastable nitrogen molecules formed in the plasma discharge. Their presence as a result of nitrogen atom recombination is well understood and documented, but their role in the subsequent chemistry is uncertain (McTaggart, 1967, Fontijn et al., 1964). The study by Clements et al. (1984), in which nitric oxide producing reactions were evident at near-ambient temperatures far downstream of the igniter, when combined with our nitrogen atom results, strongly suggests that metastable nitrogen is important in the chemistry of nitrogen plasma jets.
Ionic species were not detected in the plume: this may be the result of difficulties in maintaining ions in the molecular beam and in focusing them into the quadrupole using the present ionizer/extractor assembly. As such our detection limit for ionic species is uncertain.

CONCLUSIONS

Molecular beam mass spectrometry has been used to determine temperature and species in the discharge plume of a pulsed plasma jet igniter in a nitrogen environment at one atmosphere pressure. Laser schlieren photographs of the plume/sampling cone interaction reveal that the sampling process has negligible effect on the plume up to the time of impingement, and temperature and species measurements indicate that at later times the gas is sampled from within the plume. The temperature ranged from 50,000K to ambient, with large fluctuations found early in the discharge. Nitrogen atom concentrations were found to range from 5%, 2mm from the igniter, to less than 100ppm at 14mm.
ACKNOWLEDGEMENT

The authors thank Mr. K. Hom for his help in all aspects of the experimental work. This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Engineering, Mathematics and Geosciences Division, and the Assistant Secretary for Conservation and Renewable Energy, Office of Transportation Programs, Division of Transportation Energy of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.
REFERENCES


FIGURE CAPTIONS

Figure 1: Schematic diagram of the pulsed plasma jet igniter and sampling cone of the molecular beam system.

Figure 2: Laser schlieren photographs of the igniter/cone interaction - early time sequence.

Figure 3: Laser schlieren photographs of the igniter/cone interaction - late time sequence.

Figure 4: Jet plume and blast wave propagation at various igniter/cone distances (x - infinite, Δ - 13mm, □ - 7mm).

Figure 5: Displacement vs. time diagram for sample flow through the molecular beam system. Part (a) depicts the continuous sampling while part (b) depicts chopped beam conditions.

Figure 6: Experimental records of the N₂ and N signals from the plasma jet igniter at 2mm separation distance.

Figure 7: Molecular nitrogen TOF data. Triangles denote the time of jet firing.

Figure 8: Calculated temperatures in the plasma plume. The dashed line indicates the extent of plume penetration.

Figure 9: Plume temperature profile constructed from the time of arrival data (figure 9) for 2mm separation distance.

Figure 10: Experimental and simulated N₂ TOF curves for the plasma plume at 2mm separation distance.

Figure 11: Nitrogen atom concentration as a function of time and separation distance.
Fig. 5
Fig. 8
This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Department of Energy to the exclusion of others that may be suitable.