Studies of C$_2$H$_6$/ air and C$_3$H$_8$/ air
Plasma assisted combustion kinetics in a nanosecond discharge

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The paper presents the studies of ethane and propane/air plasma assisted combustion at a pressure of 60 torr and temperature 300K. O atoms in the plasma have been measured as a function of time after a single discharge pulse using TALIF (Two photon laser induced fluorescence) at 60 torr and temperature of 300 K for these mixtures at various equivalence ratios. A plasma chemistry model of hydrocarbons has been developed. This is done by combining a reduced mechanism of the latest low temperature hydrocarbon mechanism with plasma air chemistry along with plasma and flame NO formation chemistry. The reaction rates of excited nitrogen species with hydrocarbons are not known accurately and have not been included. O atoms measured in air are compared with the new mechanism predictions. The O atom measurements compare well with the mechanism predictions. The O atoms decay slightly faster in the case of ethane than predicted. For the case of propane, the O atoms decay much faster than predicted by the mechanism. A better mechanism for low temperature hydrocarbon combustion is required for the plasma. The O atoms begin chain reactions giving rise to OH and H radicals. H$_2$O is formed during these chain reactions. But soon all the radicals decay in ~1.5msec. After the end of chain reactions, species such as CH$_2$O, CH$_3$, C$_2$H$_4$, H$_2$O$_2$, O$_3$ begin to accumulate. CO and CO$_2$ are formed only at the end of these chain reactions through slow oxidation. The mechanism has been used to study ignition by a single discharge for air/fuel mixtures at high temperatures in the range 500-700K and pressures in the range 300-500 torr. An initial mole fraction of O atoms (~5 x 10$^{-5}$) has been added without a nanosecond discharge and the mixture is observed in time. It is found that for higher pressures of ~ 400torr, 700K, there is a two stage ignition in ~28msec for stoichiometric air while it is 43msec for same mixture at 300 torr showing that there is a dependence on initial pressure and temperature to be studied to take advantage of the nanosecond discharge.

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I. Introduction

There have been consistent efforts for decades in developing more efficient combustion for high speed aviation [1-5]. Development and successful running of hydrogen fuelled scramjets has been the pinnacle of present high speed aviation. Such propulsion systems must accelerate from zero speeds and reach high Mach numbers. This necessitates their operability at a wide range of speeds and atmospheric pressure conditions. Air breathing engines have proved to be the most efficient for supersonic systems. With regard to the development of combustion methods for such propulsion systems, we have seen different approaches, such as, subsonic combustion and supersonic combustion for hypersonic aviation. The free stream atmospheric air is compressed to supersonic speeds in scramjets to higher static pressures and temperatures and mixed with another stream of gaseous fuel flowing in parallel. The temperature and pressures of the condition of mixing of fuel and air, dictate the process of induction and combustion. The diffusive mixing of fuel and air and combustion at supersonic speeds is very complicated. It is also important to note that very low pressures cannot be adopted for supersonic combustion and propulsion as it is inefficient. This is because; the heat release is consumed in dissociating molecules into radicals at low pressure, decreasing the sensible entropy of the gas available for useful propulsion.

At low Mach numbers, addition of heat at supersonic speeds is not the most efficient means. However, at high Mach numbers, this is more efficient than the subsonic combustion adopted in ramjets. In ramjets, the static pressures and temperatures are so high that the fuel/air mixture does not proceed towards combustion. Most of the combustion takes place during the expansion in the nozzle section where the pressures and temperatures are lower. Handling of high temperatures and pressures require heavy complicated and unwanted machinery. However, the ramjets are better equipped to work at lower Mach numbers despite the fact that it requires complicated adjustments in the flow section according to the flow Mach number.

In supersonic combustion, the time available for burning of fuel is however, very less. The static pressures and temperatures are also lower. The better efficiency of the supersonic combustion method has led to lot of research in combustion possibilities in a wide range of Mach flow, atmospheric pressure conditions and fuels other than hydrogen. Plasma assisted combustion (PAC) is one method where there has been vigorous research recently, particularly with the nanosecond high voltage electric discharge method [6-21].

Here, a high voltage is applied to the fuel/air mixture or a mixing gas for a short time of the order of tens of nanoseconds to produce diffuse large volume plasma. Longer discharge times can produce arcing and instabilities. The large volume low temperature and pressure plasma of fuel/air mixtures has been found to increase the fuel burn fractions to up to 80% even under lean equivalence ratios. This technology of ignition and combustion is therefore seen to be promising for supersonic combustion at a wide range of operating conditions and for relighting combustion.

The nanosecond discharge of air/fuel can produce a wide range of radical pools such as O atoms electronically excited molecules such as N$_2$(A,B,C), singlet delta oxygen, ozone etc which can initiate ignition and combustion under adverse conditions. For this reason, plasma assisted combustion (PAC) kinetics has been studied in shock tubes [6,7] plasma flow reactors [8-11] gliding arc [14, 15], counterflow burners [16], IC engines [17] and supersonic ducts [5] with various hydrocarbon fuels C$_n$H$_{2n+2}$, $n = 1$–5 and hydrogen. The dynamics of nanosecond discharge has been studied by [19]. The diffuse plasma generates several important radicals which can initiate ignition and combustion. Microwave discharge is found to enhance flame speeds of methane/air mixtures at low equivalence ratios and temperatures [22].

It has been observed that O atoms formed by electron impact dissociation and exchange of energy with excited nitrogen molecules, initiate the ignition and combustion reactions in PAC [8-12] at 60 torr, 300K. NO molecules have also shown to be important in PAC [8, 14, 15]. Singlet delta oxygen molecules and ozone effects on flame propagation have been investigated by [13,14]. Reduction of ignition delay has been observed in shock tubes at relatively low pressures of ~0.5 atm by as high as
600K [6]. The discharge was found to be non-uniform at pressures greater than ~1.5 atm. In IC engines, nanosecond discharge has been found to be a better, faster means of depositing energy, forming ignition radicals and also developing multiple ignition kernels [17]. Ref [16] found increase in extinction coefficients of counter-flow flames when the oxidizer side is subject to continuous nanosecond discharge plasma.

However, there have not been many studies on kinetics of hydrocarbon fuels such as ethane and propane. These fuels are easily available, have high energy density and are easy to carry on flights. There have been no studies of the kinetics of reactions through measurements of species at pressures other 60 torr and 300K. The present hydrocarbon reaction mechanisms have been validated in temperatures above ~600K. The knowledge gap of plasma kinetics of hydrocarbon fuel/air mixtures in the temperature range 300-600K needs to be filled and at considerable pressures higher than 60torr pertaining to actual application conditions. However, it is difficult to generate diffuse plasmas for kinetics studies at high pressure and low temperatures. The fuel/air mixtures can only be heated close to the self ignition temperature at a particular pressure and electric discharge applied for kinetics studies. Beyond this temperature, the mixture will self ignite and plasma becomes immaterial. In this paper, the studies have been conducted upto pressures of 200torr.

From the discussion above, it is seen that there has not been much research on the plasma kinetics of simple hydrocarbons such as ethane and propane by actual measurement of species in the plasma in the temperature range of 300-600K and also at pressures considerably higher than 60torr. This is the objective of this paper. A hydrocarbon mechanism has been build for ethane and propane using low temperature chemistry of Healy et al. [26,28] butane mechanism. The mechanism has been reduced to a smaller mechanism using pathway analysis method [29, 30]. This mechanism has been incorporated into the air plasma mechanism [7,8]. NO formation has also been included based on [9, 8]. O atom density profiles have been measured at 60torr and 200torr (300K) as a function of time and compared with the mechanism predictions. Section III explains the experimental details of TALIF (two photon laser induced fluorescence) used to measure O atoms and the quartz plasma reactor assembly. Section IV explains the details of numerical simulation of the plasma and the incorporation of hydrocarbon mechanism with plasma mechanism. Section V presents the results and discussion, the summary of which is presented in section VI.
III. Experimental setup

The experimental setup consists of a quartz reactor (figure 1). The overall length of the reactor is approximately 14”. The gases are flown through a rectangular tube 10mm X 18mm outer dimensions. The thickness of the quartz walls are 1mm. The electrodes made of thin copper foil (~0.01mm thick) of dimensions 16mm x 45mm are placed outside this rectangular section. The electrodes are then sandwiched between ceramic plates after covering the electrodes with a high temperature insulation material. The quartz reactor is covered by heating tapes and then properly insulated with high temperature insulation on the air inlet side. This way gas flow inside can be heated up to ~850K. However, the gases are heated to 620±5K for the high temperature experiments. The temperature at the center of the plasma region can be measured using a 1/16” thermocouple probe when the plasma is off. Air and the fuels are mixed before reaching the plasma. The flow velocities are maintained at ~1m/s. This way the residence time of gases in the plasma is ~ (45mm/1000mm/s) = 45milliseconds. The O atoms in the plasma were measured at 300K.

The pulser was run in two modes: (1)high voltage for O atom measurements and (2)low voltage for continuous discharge and GC analysis of combustion products. Fig. 2 shows the measured Voltage-Current (V-I) characteristics of the nanosecond discharge pulser in the high voltage mode. It is seen that the discharge duration is ~12ns and the peak voltage is ~18kV. In the high voltage mode, the pulser is run at 10Hz and O atoms are measured in the plasma as a function of time. In the low voltage mode, the peak voltage is maintained at ~12kV and run at high frequencies (0.5-10kHz repetition rate). This was done to reduce EMI (Electromagnetic interference) with measuring instruments and also to prevent heating up of the pulse generator. Figure 4 shows the actual picture of the plasma at 5kHz and also a single pulse ICCD image of the plasma. From the ICCD image, it is seen that the plasma is quite uniform. All the connecting cables were sheathed and grounded to reduce EMI. The
gas flow is controlled using mass flow controllers and is controlled within 3% of the set mass flow rate values.

Figure 3: Picture of plasma at 5 kHz, 50 torr air at room temperature and ICCD image of a single pulse (inset) using a gate width of 20ns.

Figure 4: Schematic of laser setup for TALIF measurements
A. O atom density measurements by TALIF.
A plasma discharge in air, air-fuel mixtures can generate electrons, ions, atomic oxygen, \( \text{O}_3 \), electronically and vibrationally excited \( \text{O}_2 \), and excited \( \text{N}_2 \) (A,B,C). In our experiments, the focus is on the role of atomic oxygen, which is one of the most important radicals produced by nano-second plasma.

The schematic of the TALIF system is shown in figure 4. An Nd:YAG laser was used to generate 532 nm radiation to pump a tunable dye laser that was tuned to 573 nm. To produce the required 226-nm radiation, this 573 nm beam was frequency doubled and then frequency mixed with the 1064 nm output of the Nd:YAG laser. Atomic oxygen was measured at the center of the plasma. The 10-\( \mu \)J/pulse UV beam was focused 30 mm ahead of the probe volume (the center of the plasma) using a 300 mm focal length lens, to avoid saturation. The gap between the parallel electrode plates is 8 mm. The signal was averaged over a distance of \( \sim 5 \) mm. The dye laser was scanned in wavelength to characterize the atomic oxygen fluorescence spectrum. The two fluorescence signal from O atoms was collected in the direction perpendicular to the probe laser beam using two 1” quartz lens of focal length 50mm. The TALIF fluorescence was observed using an 850 nm bandpass filter of 40 nm FWHM and a Hamamatsu photomultiplier (PMT-R7154). A slit of 5mm was used on the PMT window. The dye laser was scanned across the fluorescence spectrum of O atoms and the profile recorded on a computer using SRS272 software and Stanford Research Systems boxcar. A photodiode received partially reflected signals of probe UV beam. This was used to normalize the TALIF signals. The area below the recorded fluorescence spectrum is used as a measure of O atom number density levels. The O atom number densities have been calibrated here using TALIF of xenon atoms for the case of air flow at 60 torr and found using the equation,

\[
N_o = \frac{S_o}{S_{\text{Xe}}} g_{\text{ND}} \frac{a_{21}(\text{Xe})}{a_{21}(\text{O})} \left( \frac{\sigma^2(\text{Xe})}{\sigma^2(\text{O})} \right) \left( \frac{\nu_0}{\nu_{\text{Xe}}} \right)^2 \frac{1}{F_{\text{Xe}}(T)} N_{\text{Xe}}
\]

Here, \( S_o \) and \( S_{\text{Xe}} \) are the integrated TALIF fluorescence signals of O and Xe atoms. \( a_{21} \) are the branching ratios for the atoms in discussion here. \( \sigma^2 \) is the two photon absorption coefficient. \( \nu \) is the two photon absorption wavelength of the atoms. \( F_{\text{Xe}}(T) \) is the Boltzmann distribution of O atoms in the 2P state of the three triplet ground states. This is \( \sim 0.74 \) at 300K. \( N_{\text{Xe}} \) is the number density of Xe atoms during calibration = \( P_{\text{Xe}}/kT \). Where \( P_{\text{Xe}} \) is the pressure in the reactor filled with pure xenon at temperature T. \( k \) is the Boltzmann constant. \( g_{\text{ND}} \) is the transmissivity of the neutral density filters used while recording the fluorescence spectrum of xenon which is usually three orders of magnitude higher than that of O atoms at a pressure of 60 torr. Further details are explained in \[8, 9\].

IV Numerical modeling
The basic plasma kinetic code used is that of OSU NETL group \[8-10\]. The air plasma model has been augmented with the latest low temperature C4 hydrocarbon mechanism \[26\]. The basic equations of this numerical model are described in the following sections. The electron impact cross-sections for the hydrocarbons have been included.

The plasma produces many radicals and electronically and vibrationally excited species such as O(P), O(D), \( \text{N}_2\text{(A,B,C)} \), \( \text{N}_2(\text{v}) \), N, \( \text{O}_2(\text{b}) \), \( \text{O}_2(\text{A}) \) and other charged species. These species have been included in the reactions except the charged species, which are of assumed to be of low number density or assumed to recombine and neutralize on a nanosecond time scale. A zero dimensional model has been used for the flow system. The Poisson equation for the charge and electric field in the plasma has not been used. Instead, a Gaussian voltage with a FWHM (full width half maximum) of 12nanoseconds has been used. The peak voltage is adjusted to fit the O atom number density measured. The input pulse energy is found to be close to that measured using the measured V-I (voltage-current) characteristics (figure 1) on an oscilloscope at a pressure of 60torr. Similarly, the current is also assumed to have a Gaussian profile with a same FWHM as the applied voltage profile. The peak
current can also be varied to match the applied pulse energy but is assumed to be close to that measured on the oscilloscope. These current and voltage values are used for the electron Boltzmann equation solutions and to obtain the rates for the electron impact processes. The EEDF (electron energy distribution function) is obtained by solving the electron Boltzmann equation which is expanded using 2 term expansion of the spherical harmonics. The electron impact cross sections are taken from literature for air and also the fuels: ethane and propane. All the electron impact products of the fuels which include H, H2, CH, C2, C2H, C2H2, C3H, CH3, CH2, for ethane and additional fragments C2H7, C3H6, C3H5, C3H4, C3H3, C3H2 for the electron impact dissociation of propane has been included in the plasma mechanism [27].

The low temperature hydrocarbon mechanism is that of NUI Galway [26,28]. The butane mechanism has been reduced using PFA (path flux analysis) method developed [29, 30]. The reduced hydrocarbon mechanism has 647 reactions and 102 species. The C4 species have been removed and species only up to C3 have been used. This has been added to the air plasma mechanism [7, 8] along with the electron impact cross sections for the fuels already mentioned. The plasma NO mechanism (N2(v) + O→NO + N) along with that of GRI has also been included. This is particularly important in view of the reaction NO + HO2 → OH + NO2 which can enhance ignition and combustion at low temperatures.

V. Results

A. O atom measurements using TALIF

O atoms were measured as a function of time under the following flow conditions
(1) Air (60torr, 200torr pressures at 300K)
(2) Air+Ethane (Φ=0.22, P=60torr, 200torr, T=300K)
(3) Air+Propane (Φ=0.11, P=60torr, 200torr, T=300K)

Figure 4: O atom number density compared with mechanism predictions in dry air at 300K, 60torr total pressure.

Figure 4 shows the measurements of O atom number density in air compared with the calculation using the new C3 plasma mechanism. The predictions are within ~10% of the measured O atom
densities. Here, the input pulse energy has been adjusted to 1.1mJ/pulse to get the required peak O atom densities measured by the experiments. The measured pulse energy is ~ 1.25mJ/pulse using the voltage-current profile on an oscilloscope (figure 2). The peak voltage is taken to be 9.635kV across a distance of 10mm. Assumed peak current is 8.5A.

Figure 5 shows the normalized O atom number density measured for ethane compared with mechanism predictions. It is seen that the measured O atoms decay somewhat faster than in air. However, the mechanism does not predict much change compared with air.
Figure 6 shows the normalized O atom number density measured for propane compared with mechanism predictions. It is seen that the measured O atoms decay much faster than in air. However, the mechanism does not predict the O atom decay so well. The O atoms possibly react much faster with the propane molecules than in the mechanism.

The LHV (lower heating value) of propane is ~ 40% higher than that of ethane (1.43 MJ/mole for ethane, 2.04 MJ/mole for propane). However, it is found from calculations that the net exothermic
heat release from propane in the plasma, is approximately double that of ethane for the same calorific heat flow of the fuels with air (with $\Phi$=1 for ethane and propane in this case) at flow conditions of 500K, 100torr total pressure. The measured input energy is $\sim$1.1mJ/pulse. The thermalized energy for ethane is $\sim$1.2mJ/pulse at the end of $\sim$100msec. The thermalized energy for propane is $\sim$1.72mJ/pulse. This is due to the more number of faster reactions of O atoms and chain propagation with propane than ethane.

For example from the mechanism, at 500K and 100torr total pressure, the possible reactions of O atoms with the fuels are,

\[
\begin{align*}
\text{C}_2\text{H}_6 + O &= \text{C}_2\text{H}_5 + \text{OH} & k &= 9.2 \times 10^7 \text{ cm}^3/\text{mole/s} \\
\text{C}_3\text{H}_8 + O &= \text{IC}_3\text{H}_7 + \text{OH} & k &= 5.75 \times 10^9 \text{ cm}^3/\text{mole/s} \\
\text{C}_3\text{H}_8 + O &= \text{NC}_3\text{H}_7 + \text{OH} & k &= 1.84 \times 10^8 \text{ cm}^3/\text{mole/s}
\end{align*}
\]

Figure 8(a): The mechanism predictions of the evolution of species after a single discharge pulse.
Figure 8(b): The mechanism predictions of the evolution of species after a single discharge pulse.

Figure 8 shows the evolution of species after a single discharge pulse. As the O atoms are formed, chain reactions develop and OH is formed. There is formation of radicals C2H₅ at first due to H abstraction reactions to form OH radicals.

\[ \text{C}_2\text{H}_6 + \text{O} \rightarrow \text{C}_2\text{H}_5 + \text{OH}. \]

H₂O is formed rapidly in the beginning but not CO or CO₂. As the O atoms die down, there is accumulation species such as C₂H₅, CH₄, and CH₂O. The formation of CO and CO₂ occurs after the O atoms die down, mostly due to slow oxidation of fuel reaction fragments with HO₂ and O₃. Similar trends are observed in propane/air flow. The calculations show N₂(A) exists for ~10 µsec. Reactions of N₂(A) with fuel and their rates are not known and have not been included in the model. However, it is known that N₂(A) can easily react with fuels such as CH₄ and dissociate them into fragments. It is possible that it is contributing to the dissociation of propane molecules, the products of which react with O atoms initially, possibly accounting for the faster decay of O atoms measured. From these figures it is seen that the initial time of ~10µsec marks the chain reaction period due to O atoms and N₂(A) molecules. It has been shown that in a 60torr, 300K flow of air, NO can be formed in a time scale of ~100µsec. The positive effect of the presence of NO in low temperature is well known due to the reaction

\[ \text{NO} + \text{HO}_2 \rightarrow \text{NO}_2 + \text{OH}. \]

This reaction has been included in our mechanism. The reaction rates of vibrationally excited N₂(v) with O atoms to form NO is not yet know accurately. These factors are important in a supersonic combustion.

From an analysis of figure 8 of the combustion of ethane, we see that ethane can react with O atoms to give rise to OH, H and C₂H₅ radicals. The O atoms are both formed and consumed. Some reactions give rise to OH and H radicals. But other reactions go into formation of oxygenated hydrocarbons and inactive species such as O₃, HO₂ and H₂O₂. The fuels react by forming intermediates such as C₂H₆, C₂H₄, CH₂O, H₂ etc. Once the O atoms die down, these species begin to accumulate.
The mechanism was used to study the ignition and combustion abilities of nanosecond discharge at different pressures and start temperature of 500K. The mechanism was run with some O atoms to begin with and tested for ignition and combustion. The O atoms in the beginning are taken to be a mole fraction of 5e-5 in an air fuel mixture at equivalence ratio, Φ=1. It is found that the initial O atoms started chain reactions giving rise to radicals such as H and OH. But the O atoms decayed in time of the order of 1.5msec for pressure range of 60-400torr. Species such as CH₄, C₂H₆, HO₂, H₂O₂ begin to accumulate at the end of these chain reactions. The increase in gas temperature was only ~12K in this period. The gas temperature was further increased to 600K and a total pressure in the range 400-600torr. For the case of 700K, 400torr propane/air (Φ=1), a two stage ignition was observed. The O atoms decayed in the initial phase of ~1.5ms. The OH radicals decay in this time but begin to increase afterwards leading to an ignition of the mixture and a sustained combustion. There is rapid ignition after 28msec and the temperature reaches adiabatic flame temperature. For the case of 700K, 300torr propane/air (Φ=1) flow, the ignition delay is found to be 43msec. These studies point to the importance of higher pressures and temperatures to start ignition and combustion to take advantage of nanosecond discharge. Although the question remains about how to create large volume diffuse plasma at higher pressures, these studies are important for supersonic combustion. High voltages may be required to attain large volume diffuse plasma at higher pressures.

VI. Summary and future work

O atom number densities have been measured in air and calibrated using xenon TALIF. A new mechanism for plasma combustion of ethane and propane has been developed. This was done by combining air plasma model along with NO formation mechanisms in plasma and flame. This was combined with a reduced mechanism of GUI Combustion Chemistry Centre. The mechanism predicts well the O atoms in air. O atoms decay slightly faster in ethane/air than predicted by the mechanism. However, the O atoms decay much faster in the case of propane/air compared to predictions of the mechanism. The mechanism does not include an accurate model of plasma NO formation mechanism, which is still under development. The reactions of excited nitrogen molecules N₂(A,B,C) with fuel molecules and their breakdown into fragments has not been included in the model. Oxidation reactions by O₂ have not been included. The O atoms formed by the nanosecond discharge start chain reactions forming radicals such as H, OH, C₂H₅. But soon the O atoms and OH radicals are not sustained and die down. At the end of decay of O atoms, species such as CH₂O, CH₄, C₂H₆, H₂O₂ begin to accumulate. H₂O is formed during the chain reactions. CO and CO₂ are formed at the end of the decay of O atoms and chain reactions through slow oxidation by H₂O₂, HO₂.

The mechanism has been used to study possible ignition at higher pressures and temperatures of air/fuel. It is found that propane/air mixture can ignite at higher pressures approximately greater than 500torr, and initial temperatures in the range of 600K when an initial mole fraction of 5 x 10⁻⁵ was assumed for the O atoms with no subsequent plasma discharge applied. Our future plan is to verify this through experiments. The experiments and calculations show that better low temperature hydrocarbon mechanisms are required. Multiple continuous plasma discharge should also be studied for plasma ignition studies. Measurements of species such as OH and sensitivity analysis can give further insight into the combustion kinetics.

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