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PLATE BOUNDARY LAYER: I. EXPERIMENTAL RESULTS

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CATALYZED COMBUSTION OF H_2 /AIR MIXTURES

IN A FLAT PLATE BOUNDARY LAYER:

I. EXPERIMENTAL RESULTS

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CATALYZED COMBUSTION OF H_2 /AIR MIXTURES

IN A FLAT PLATE BOUNDARY LAYER:

PART I. EXPERIMENTAL RESULTS

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ABSTRACT

A study has been made of the combustion characteristics of lean hydrogen-air mixtures flowing over a heated catalytic platinum plate. The objectives of the investigation were to develop a better understanding of the interaction between fluid mechanics, gas phase combustion, and surface reaction in high temperature surface catalyzed combustion. The experimental system consisted of a thin quartz plate with vacuum deposited platinum heating strips mounted over an open atmospheric pressure jet of premixed hydrogen and air. Boundary layer density profiles were measured using differential interferometry for flow visualization studies and Rayleigh scattering for point density measurements. Laser Doppler velocimetry was also used to measure the velocity distribution. The presence of heat release due to surface reaction was determined from changes in heating strip power inputs with fuel addition. Results were obtained for a range of equivalence ratios from 0.05 to 0.3 and plate surface temperatures from 470K to 1300K. Significant surface heat release was found for all mixtures at plate temperatures as low as 470K. At increased equivalence ratios and plate temperatures heat

release due to gas phase reaction was present. This was characterized by local increases in temperature across the boundary layer and an increase in thermal boundary layer thickness. Comparisons with numerical calculations based on a surface reaction model which included finite rate surface oxidation of H_2 and radical recombination indicated qualitative agreement in general boundary layer behavior, but the predicted gas phase heat release rate was considerably higher than found experimentally.

1. INTRODUCTION

Interest in the use of catalytic surfaces to promote combustion reactions has increased greatly over the past several years. A number of studies have been undertaken which indicate that catalytically supported combustion makes efficient burning of a variety of fuels possible under fuel lean conditions with a substantial reduction in NO_x levels.[1,2] While initial emphasis was based on the use of catalytic combustors for aircraft gas turbine application [1,3] its potential application to stationary power sources has also been studied with favorable results.[4]

Much of the work to date has involved parametric investigations of prototype catalytic combustor configurations. Such work is necessary to the development of practical catalytic combustors and the results have been quite promising. However, such studies are somewhat limited in terms of deriving a more fundamental understanding of catalytically supported combustion. Two such areas where a greater knowledge would be desirable are the role of internal heat and mass transfer in the catalytic combustion process, and the role of homogeneous as opposed to catalytic surface reactions. An understanding of these and related processes such as pollutant generation is necessary for the optimum evaluation and application of the surface catalysis concept to practical combustion system design.

The approach of the present investigation has been to examine catalytically supported combustion in a well characterized system in which most of the important physical and chemical processes found in larger scale catalytic combustors are present. The geometry chosen consists of combustion in the boundary layer of heated catalytic flat

plate. This system provides a suitable geometry for both experimental and numerical modeling studies. The operation of such a system under a suitably selected range of conditions greatly facilitates determination of the roles of the various processes involved. The parallel development of a numerical modeling program was necessary for the analysis of the experimental results.

The experimental study has concentrated on the use of three optical diagnostic techniques for boundary layer measurements. These are laser Doppler velocimetry, Rayleigh scattering, and differential interferometry. Optical diagnostic techniques were used because they are non-perturbing to the flow and provide good time and spatial resolution. Laser Doppler velocimetry was used for the measurement of the boundary layer velocity distribution primarily to determine if the experimental system does provide a good approximation to an idealized flat plate flow. Both differential interferometry and Rayleigh scattering were used to obtain boundary layer density profiles. The advantage of a flow visualization method such as interferometry is that a relatively rapid study of boundary layer behavior under combustion conditions is possible. Rayleigh scattering was then used for a more detailed investigation of the boundary layer structure under those conditions of greatest interest.

In the remainder of this paper the experimental facility and diagnostics are described, and results are presented for the flow of chemically reacting H_2 /air mixtures over a heated platinum catalytic surface. The experimental results are compared with numerical calculations based on a detailed kinetic mechanism for gas phase reactions and a simplified

model for surface reaction which includes finite rate surface oxidation of H_2 to H_2O and radical recombination. Details of the numerical calculations and the effect of various surface reaction models are presented in the following paper [5], hereafter referred to as II.

2. EXPERIMENTAL

2.1 Flow System and Plate Design

The experimental configuration consists of a thin quartz plate with vacuum deposited platinum heating strips suspended vertically over an open, atmospheric pressure jet of premixed hydrogen and air. The jet is produced by a stagnation chamber 20 cm in diameter and fitted with a 5 cm diameter converging nozzle at the exit. It has three internal screens and produces a uniform, low turbulence flow of air or premixed combustible mixture which is directed vertically upwards. Lower gas flow rates are metered by calibrated rotameters, and larger air flow rates are metered using a standard orifice gauge and water manometer. The house air supply is passed through a dryer and filter combination which eliminates most particulate matter. The flat plate and stagnation chamber are mounted on a three axis traverse mechanism with a 0.001 cm positioning sensitivity.

Details of the flat plate and holder design are shown in Fig. 1. The plate is made from a 1.5 mm thick, 75 mm square quartz sheet with a sharpened leading edge angle of 2° . Five platinum heating strips are vacuum deposited on the plate surface, oriented perpendicular to the flow and of varying widths to improve temperature control near the plate leading edge. The central region of the strips is coated to a thickness of 0.3 microns, while the outer edges are coated to approximately three times this thickness. This keeps the outer edges of the plate sufficiently cool so that gold foil can be used to make electrical contact with the stainless steel fingers. Electrical power is individually controlled to each strip and as a result a reasonably uniform plate surface temperature

can be maintained.

To reduce the interference of surface scattering of the laser beam with Rayleigh scattering in the boundary layer, important when measurements are being made close to the plate, it has been found advantageous to have a small bend, about 2° , in the center of the plate. The Rayleigh scattering measurements are made in the shadow of this bend. This results in a substantial reduction in plate scattered light and makes measurements possible as close as 0.2 mm from the surface.

Surface temperatures greater than 1000°K are measured using a disappearing filament type optical pyrometer and an emissivity estimate of 0.65 for the platinum surface. Surface temperatures below 1000 K are determined from the measured resistivity of the platinum strips.

2.2 Optical Diagnostics

2.2.1 Laser Doppler Velocimeter

The optical arrangement for the laser Doppler velocimeter and Rayleigh scattering systems is shown in Fig. 2. A Spectra-Physics argon-ion laser operating at 488 nm with 1 watt power is used in both systems. The laser Doppler velocimeter is of the intersecting dual-beam type with real fringes [6]. A cube type beam splitter (not shown in the figure) consisting of two 90° equilateral prisms cemented along the hypoteneuse is used. This splits the laser beam into two beams with equal optical path length and variable beam separation. The two beams are focussed by a lens and intersect at the measurement volume. Scattering from the intersection region is collected at a 30° angle from the forward direction by a 55 mm focal length camera lens which

images a pinhole on the intersection region, and the signal is detected with an RCA type 931 A photomultiplier. The frequency of the bursts is determined by a Thermal Systems Model 1090 frequency tracker.

Aluminum oxide particles of nominally 2.0 micron diameter are introduced into the air supply prior to the stagnation chamber. The particles are suspended in water and dispersed into droplets by a collision type atomizer.

2.2.2 Rayleigh Scattering

Rayleigh scattering refers to the elastic scattering of light from gas molecules [7]. The intensity is proportional to the gas density and gives approximately the same information as an interferometric measurement of gas density. The principal advantages over interferometric techniques [8] is that the density at a single point is measured rather than integrated along the optical path. A further advantage is that with moderately powered lasers the intensity is sufficient to provide adequate time resolution and signal-to-noise ratio for studying turbulent reacting flows [9].

The Rayleigh scattering is measured at right angles to the focussed laser beam by a monochromator and photomultiplier. A waist diameter of 50 microns was chosen to give good spatial resolution in the boundary layer. The scattered light is collected at an angle of 90° to the incident beam by an $f/1.2$, 55 mm focal length camera lens. An image of the monochromator slit, 100 microns wide and 1 mm long, is then focused on the waist region of the laser beam to define the measurement volume. The optics and monochromator (a 0.3 m McPherson) are arranged

such that all the rays from the lens fall onto the photomultiplier, an RCA type 1P28. The output current of the photomultiplier is measured using a Keithley electrometer. For this system the expected photoelectron counting rate can be estimated [9] and should be about 10^7 counts/s for air at room temperature. The observed signal-to-noise ratio is consistent with this signal level. To minimize surface scattering a series of spatial filters are used to "clean up" the laser beam in addition to the bend on the center of the plate.

2.2.3 Differential Interferometer

To study the changes in the flow pattern as a function of variations in operating parameters, differential interferometry was used to visualize the flow field. This technique is based on the interference of two beams passing through adjacent points in the test space. Creeden [10] has shown that in this situation, the fringe displacement is proportional to the derivative of the index of refraction, rather than to the index of refraction as in the case of a conventional interferometer. Since the index of refraction is linearly proportional to the density, the fringe displacement is proportional to the density gradient in the test space provided that all other index of refraction fields along the optical path are negligible.

A schematic of the optical arrangement is shown in Fig. 3. The laser beam is first expanded by a short focal length lens, L1 ($f = 15$ mm). A second lens, L2 ($f = 125$ mm), is placed confocally with L1 to produce a parallel beam about 5 cm in diameter. This passes through the test region which is in turn imaged onto a ground glass screen by L3

($f = 1000$ mm). A microscope slide is placed near the focus of L3 at a large inclination so that multiple reflections from the surfaces of the slide give rise to the interfering beams. The interferogram formed on the screen can be observed visually and photographed with a 35 mm camera.

An example of an interferogram is shown in Fig. 4 for a plate surface temperature of 1200 K and an equivalence ratio of 0.3. The distribution of the gradient of the refractive index in the boundary layer is shown by the displacement of the fringes from their undisturbed position (found by extrapolation of the fringes outside the boundary layer in the free stream).

To obtain density or temperature profiles across the boundary layer, the fringe displacement is measured directly from the interferograms. Numerical integration of the area under the fringe displacement curves then gives the difference between the density of the gases in the free stream (ρ_∞), and the local gas density. The corresponding temperature profiles are obtained using the ideal gas law. Details of the analysis procedure can be found in ref. 11.

3. RESULTS AND DISCUSSION

3.1 No Combustion

A series of initial measurements were taken in the boundary layer of an unheated plate and a heated plate with no fuel addition to characterize the flowfield and verify the numerical calculations. Velocity measurements in the case of the unheated plate showed excellent agreement with the Blasius profile [12]. Velocity profiles for a heated plate are shown in Fig. 5 for distances of $x = 5$ mm and 20 mm downstream of the plate leading edge. The results presented are for an average plate surface temperature of 1180°K and a free stream velocity of 3.17 m/s. The solid lines represent the results of numerical calculations based on the computer program described in II [5]. Agreement between the experimental and predicted profiles is within 3%.

When the plate was heated it was found that the particles added to the flow for the LDV measurements moved away from the surface of the plate. Thus, as can be seen in Fig. 5, no velocity measurements could be made in the region of the thermal boundary layer closest to the plate. A detailed investigation, both experimental and computational, of the particle motion has been carried out and it appears that the particle motion is due to a thermophoresis effect. Preliminary results can be found in [13]; a final publication is in preparation.

Rayleigh scattering density measurements were used to calculate the boundary layer temperature profiles. Typical results are shown in Fig. 6 for a plate temperature of 1200°K and distances of 5 mm and 20 mm downstream. These measurements were corrected for background surface scattering, which contributed about 10% of the signal at 0.5 mm

from the surface. Agreement with the numerical computations is again quite good. Based on a comparison between indicated surface temperature using the optical pyrometer and extrapolation of the boundary layer temperature profiles to the plate surface, an emissivity estimate of 0.65 was made for the plate surface. This value was used in all subsequent surface temperature measurements using the optical pyrometer.

From the above measurements and comparison with theory it was concluded that the experimental design provided a good approximation to idealized flat plate flow, that the laser Doppler velocimeter measurements are satisfactory, and that the measurement of density by Rayleigh scattering is a viable technique capable of accurate results.

3.2 Boundary Layer Combustion

3.2.1 Differential Interferometer Study

Typical temperature profiles obtained from an analysis of the interferograms are shown in Fig. 7 for a plate temperature of 1270 K at a distance of 50 mm from the plate leading edge. The profiles for no fuel and for an equivalence ratio of 0.1 are typical of those with little or no heat release due to gas phase reaction. Under these conditions the profiles are approximately self-similar. As the equivalence ratio is increased the temperature profiles become significantly different. In this case the heat release rate due to gas phase combustion is sufficient to result in an increase in local gas temperature across the boundary layer and a reduced temperature gradient at the surface.

It should be noted that the temperature profiles obtained from the interferograms are only approximations to the actual profiles measured

with Rayleigh scattering and predicted by the numerical calculations. This can be attributed to experimental error, and to various assumptions made in the analysis [10]. As can be seen in Fig. 7 the interferograms do, however, provide the information needed to distinguish between various stages of gas phase reaction. Further analysis of the data over a range of conditions shows that the existence of gas phase combustion in the boundary layer depends on mixture composition, surface temperature, and distance from the leading edge.

The conditions under which various stages of gas phase combustion are present are summarized in Fig. 8. These results are based on the analysis of interferometer data taken at a distance of 50 mm downstream from the plate leading edge. At distances greater than this fluctuations in the jet mixing layer become a significant source of disturbance to the boundary layer. If the plate temperature is not sufficiently high or if the gas mixture is very lean, then the gas phase reaction rates are low and little heat release due to gas phase combustion occurs. In this case local temperature increases due to gas phase heat release are limited by the relatively rapid rates of conduction and convection of heat into the outer boundary layer and downstream regions. It is important to note, however, that significant surface reaction was present even at low temperatures and equivalence ratios. Surface reaction was measured at temperatures as low as 470 K, the minimum surface temperature at which measurements were taken. This was apparent as an increase in plate surface temperature due to surface energy release, as fuel was added to the flow. By decreasing the power input to the individual heating strips when surface

reaction was present the plate temperature could be maintained at the desired value.

At increased temperatures and equivalence ratios a region exists where both surface reaction and boundary layer combustion occur simultaneously. Under these conditions the location at which gas phase combustion occurs is a function of both plate surface temperature and equivalence ratio, and an increase in either surface temperature or equivalence ratio results in an upstream movement of the combustion zone toward the leading edge. At the highest temperatures and equivalence ratios combustion is apparent very near the plate leading edge and the primary heat release, or reaction, zone develops a flame-like structure characterized by a steep temperature gradient which extends well out into the free stream.

3.2.2 Rayleigh Scattering Study

Based on the results of the differential interferometer study a set of conditions was chosen for Rayleigh scattering measurements in which various stages of gas phase reaction were present. The set of conditions at which Rayleigh scattering measurements were taken is indicated in Fig. 8. These measurements, in addition to providing a better understanding of boundary layer behavior under combustion conditions, also provide a set of data upon which the development of the numerical model can be based.

Density profiles at a distance of 30 mm downstream from the plate leading edge are shown in Fig. 9 for a surface temperature of 1170 K and various equivalence ratios. Also shown as a solid line is the corresponding density profile obtained from numerical calculations for

the case of no combustion. At equivalence ratios less than 0.15 the experimental data are in good agreement with the calculations for no combustion present, indicating no significant heat release due to gas phase reaction. Under these conditions gases in the boundary layer are heated entirely by cross stream conduction from the high temperature plate surface. At equivalence ratios of 0.20 and 0.25 the thermal boundary layer becomes considerably thicker due to gas phase heat release and a region of approximately constant temperature exists across much of the boundary layer due to the heat release from gas phase combustion.

The effect of plate surface temperature on thermal boundary layer thickness is shown in Fig. 10 for an equivalence ratio of 0.20. The thermal boundary layer thickness, δ_T , is defined here as the point at which $\rho/\rho_\infty = 0.5$. The corresponding results from numerical calculations for the case of no combustion are indicated by solid lines. At a plate temperature of 1070 K no gas phase combustion is present and the experimental and predicted profiles are in excellent agreement. At temperatures of 1170 K and 1270 K a noticeable increase in δ_T occurs at a distance of approximately 7 mm from the leading edge due to the heat release in the boundary layer from gas phase combustion. It can be seen that the departure from predicted boundary layer thickness for no combustion is relatively gradual for a plate temperature of 1170°K. At 1270°K, however, the gas phase heat release rate is higher and a rapid increase in δ_T occurs between 15 mm and 20 mm from the leading edge. Further downstream the thermal boundary layer growth rates become comparable for the two plate temperatures.

3.3 Comparison with Numerical Calculations

In II a computer program based on a finite difference technique is developed to solve the boundary layer equations for laminar flow over a heated plate with H_2 /air combustion. Realistic transport properties and a detailed gas phase kinetic mechanism are included in the program. Three models are formulated to account for catalytic reaction at the plate surface. These are based on 1) the limiting case of an equilibrium surface at which species concentrations are driven instantaneously to their equilibrium values, 2) a boundary condition which includes finite rate surface oxidation of H_2 to H_2O , and 3) a condition which includes radical recombination at the plate surface in addition to surface oxidation of H_2 . Results are also presented for a noncatalytic surface to investigate the effect of surface reactions on the combustion process.

Because the conditions at which most of the calculations were made differ from those at which experimental results were obtained a direct comparison between the experimental results and the numerical results in II is difficult. It is possible, however, to make some qualitative observations. For an equivalence ratio of 0.1, a plate temperature of 1100 K, and a free stream velocity of 3.17 m/s the calculations for a noncatalytic plate surface predict a factor of 4 increase in δ_T over that found with no combustion present. With an equilibrium surface boundary condition (condition 1) above) the corresponding increase in δ_T is approximately a factor of 2, and when finite rate surface reaction is included in the model (conditions 2) and 3)) the predicted increase in δ_T is only 30%. Surface reaction is thus found to result in a substantial reduction in gas phase heat

release rates due to removal of H_2 in the high temperature region near the plate surface and quenching of radical species generated in the gas phase. The experimental results shown in Fig. 10 for a plate temperature of 1170 K and an equivalence ratio of 0.20 indicate an increase in δ_T of approximately 20% over the case of no combustion. Thus the above models for finite rate surface reaction provide qualitative agreement with the relatively small increase in thermal boundary layer thickness due to combustion observed experimentally.

To obtain a more direct comparison between experimental results and the numerical model, the calculations were repeated for conditions under which gas phase reaction was measured experimentally; an equivalence ratio of 0.2, a plate temperature of 1170°K, and a free stream velocity of 1.5 m/s. Since both radical recombination and surface oxidation of H_2 are likely to be important in the present system, this was chosen as the most realistic boundary condition to apply in the case of a catalytic surface. The calculated thermal boundary layer thickness is compared with the experimental results taken from Rayleigh scattering measurements in Fig. 11.

As can be seen, the experimental results indicate slower ignition of the gas phase reactions and a lower heat release rate than predicted numerically. Since good agreement was obtained between the numerical and experimental results with no combustion present, the most likely cause of the observed discrepancy appears to be due to either the model employed for surface reaction or the gas phase kinetic mechanism. The reaction rate used for surface oxidation of H_2 was based on surface reaction rates measured in the present system under comparable operating

conditions and should provide a representative rate for H_2 surface oxidation [11]. Additional calculations in which this rate was increased by a factor of 10 indicate that the onset of gas phase combustion and the corresponding increase in thermal boundary layer thickness are relatively insensitive to this rate over the range considered. A factor of 10 is somewhat greater than the expected experimental error, so it does not appear that uncertainties in the H_2 surface oxidation rate can account for the discrepancy. Since a reaction probability of unity for each collision of a radical species with the surface was assumed in the model for surface radical recombination, the computations have assumed the maximum possible surface quenching effect on gas phase reaction.

Based on the above discussion it appears that the high predicted heat release rates are due to uncertainties in the gas phase kinetic mechanism used. Although the best available values for gas phase kinetic rate constants from the literature were used, some uncertainty exists in each of these rates. This is particularly true of those reactions involving HO_2 , several of which are important during both the initial radical buildup period and in the primary heat release zone. Uncertainties in the individual reaction rate constants are currently being evaluated and an analysis will be made to determine the effect of these uncertainties on the combustion process.

4. CONCLUSIONS

A study of boundary layer combustion for lean H_2 /air mixtures flowing over a heated platinum plate indicated the presence of significant surface heat release for all equivalence ratios studied and plate surface temperatures as low as 470°K. At increased equivalence ratios and plate temperatures both surface reaction and stable boundary layer combustion were found to occur simultaneously. Boundary layer combustion was characterized by an increase in local gas temperature across the boundary layer and an increase in thermal boundary layer thickness over that obtained with no combustion. Under these conditions the location at which gas phase combustion occurred was a function of both plate surface temperature and equivalence ratio. An increase in either surface temperature or equivalence ratio resulted in upstream movement of the combustion zone toward the plate leading edge where the primary reaction zone developed a flame-like structure characterized by steep temperature gradients which extended well out into the free stream.

Numerical calculations based on a model for catalytic surface reaction described in II [5] which included finite rate surface oxidation of H_2 and surface radical recombination predicted more rapid gas phase combustion than was found experimentally. An examination of the surface reaction model indicated that uncertainties in the H_2 surface oxidation rate could not account for the observed discrepancy. The model used for surface radical recombination, in which a reaction probability of unity was assumed for each collision with the surface, should provide the maximum possible quenching effect on gas phase reaction. Thus it appears that the rapid gas phase combustion rate predicted numerically is due to uncertainties in the gas phase kinetic mechanism.

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- Fig. 4. Interferogram of the flat plate boundary layer.
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Fig. 10. Effect of plate surface temperature on thermal boundary layer thickness ($\rho/\rho_\infty = 0.5$) for flow over a heated platinum plate. H_2 /air combustion. $\phi = 0.20$, $U_\infty = 1.5$ m/s. Solid lines indicate numerical results for no combustion.

Fig. 11. Comparison of experimental and predicted thermal boundary layer thickness profiles for a heated platinum plate. H_2 /air combustion. $\phi = 0.20$, $T_s = 1170^\circ\text{K}$, $U_\infty = 1.5$ m/s.

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