THE ROLE OF SURFACE GENERATED RADICALS IN CATALYTIC COMBUSTION*

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The role of surface generated OH radicals in determining the catalytic ignition characteristics for propane oxidation on platinum is under study. The experiments are being conducted in a stacked-plate, catalyst bed. Transient measurements, during catalytic ignition, of the catalyst's axial temperature profile have been made and the effect of equivalence ratio, inlet temperature and inlet velocity are being investigated. These measurements will provide insights which will be useful in planning and interpreting to OH measurements. Attempts to measure OH concentration in the catalyst bed using resonance absorption spectroscopy have been unsuccessful, indicating that OH concentrations are below 10^{16}/cc but still possibly above equilibrium values. Measurements are currently underway using forward scatter laser induced fluorescence which should extend the OH detection limits several orders of magnitude below the equilibrium concentrations.

INTRODUCTION

The advantages of catalytic combustion over conventional combustion are lower emissions, higher efficiency, increased operational stability, stable operation at lower equivalence ratios, improved pattern factors, and wider fuel specifications. The performance of a catalytic combustor is determined by the interaction of a number of physical and chemical processes, including convection and diffusion of species, heat and momentum; conduction and radiation heat transfer in the substrate; and gas phase and surface chemical reactions. An important, and as yet unresolved, question regarding the interaction of these fundamental physical and chemical processes is the effect of catalytic surface reactions on the gas phase chemistry. In particular, it is the effect of intermediate and radical species, generated by catalytic wall reactions, on the gas phase chemistry which has not been established. That radicals can be produced by catalytic surface reactions and that at sufficiently high surface temperatures the radicals are desorbed by the surface into the gas phase has been well documented in low pressure flow experiments [1-5]. However, the importance of this phenomenon in catalytic combustion and the role of these radicals in determining the performance characteristics of catalytic combustors has not been established.

EXPERIMENTS AND STATUS

A schematic drawing of the stacked-plate, catalytic combustor in which the propane-air catalytic ignition experiments are being conducted is shown in

269

Figure 1. The catalyst plates are 50 mm wide, 100 mm long and 1 mm thick. There are a total of nine plates in the catalyst bed with an inter-plate spacing of 6 mm. The catalyst plates are made from a steel alloy substrate, coated with aluminum oxide and then platinum. Six 0.75 mm diameter holes have been drilled into the side of the center plate at six axial locations. Thermocouples have been inserted into these holes for measurement of the plate's axial temperature profile during ignition. A water cooled gas sampling probe and a thermocouple are used to obtain gas composition and temperature measurements at the exit of the catalyst bed. Optical access is available through windows in the sides of the test section located at several axial positions. The window aperture is 10 mm, therefore the entire distance between two plates can be probed.

Based on a similar experiment by Cattolica and Schefer [6] it was anticipated that the OH concentrations would be large enough for detection by resonance absorption spectroscopy. Under the conditions of this experiment the OH detection limit using this technique is about 10^{16}/cc, as compared to equilibrium OH concentrations of about 10^{14}/cc (based on the overall equivalence ratio and the catalyst temperature - see Figure 2). Measurements were attempted in the stacked-plate combustor using resonance absorption and were unsuccessful, implying that the OH concentration is less than 10^{16}/cc but still possibly greater than the overall equilibrium concentration. In order to extend the OH detection limits it was decided to use laser induced fluorescence (LIF). Because of the limited optical access with the stacked-plate configuration, it has been necessary to use forward scatter collection. The spatial resolution with forward scatter collection is comparable to that of the absorption measurements, which is adequate for the 2-D flow field between the catalyst plates. The forward scatter LIF system is currently being evaluated in a flat flame burner by comparing it to simultaneous absorption measurements, after which it will be applied to the catalytic combustor.

Although there have been several [7-9] theoretical studies of catalytic ignition there have been no experimental studies reported to date. In order to better understand the characteristics of catalytic ignition, especially for the purpose of providing insights to guide the CH experiments, it was decided to investigate the effects of equivalence ratio, inlet temperature and inlet velocity on catalytic ignition. Experiments have been conducted where the inlet temperature and velocity are fixed, the fuel is turned on and the transient response of the catalyst axial temperature profile is measured. Preliminary results from the catalytic ignition experiments are shown in Figures 3-5.

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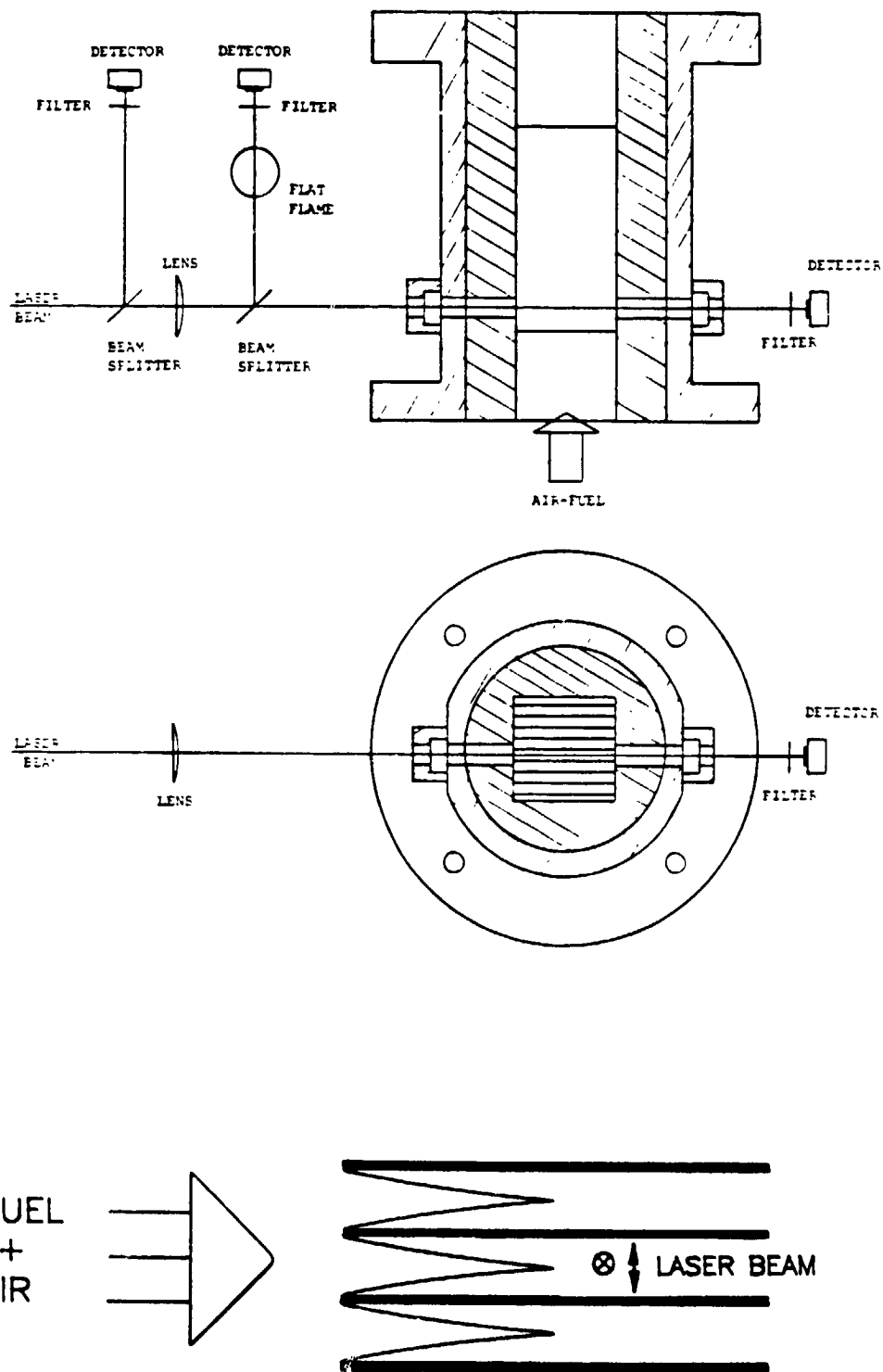
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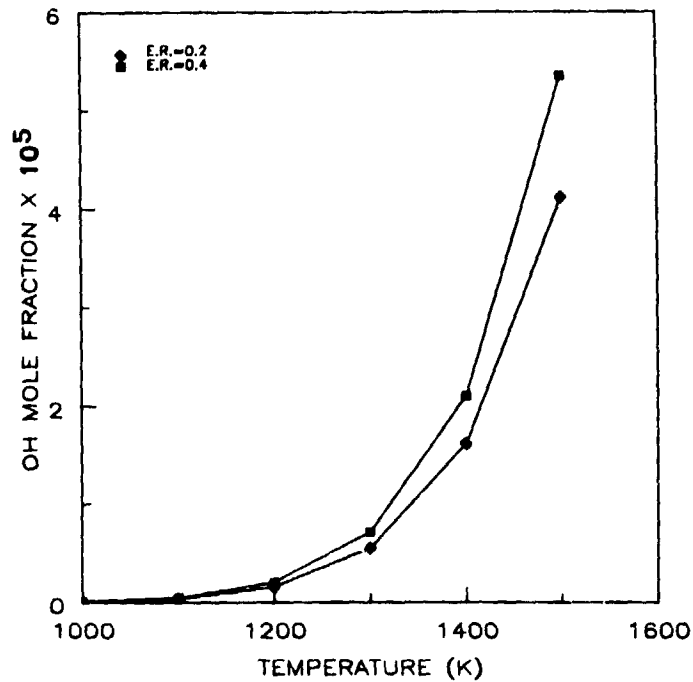Figure 1 — Schematic drawing of stacked-plate catalytic combustor with OH absorption experiment.

Figure 2 — OH equilibrium concentration versus temperature for initial equivalence ratios of 0.2 and 0.4. Calculated using NASA Equilibrium Code.
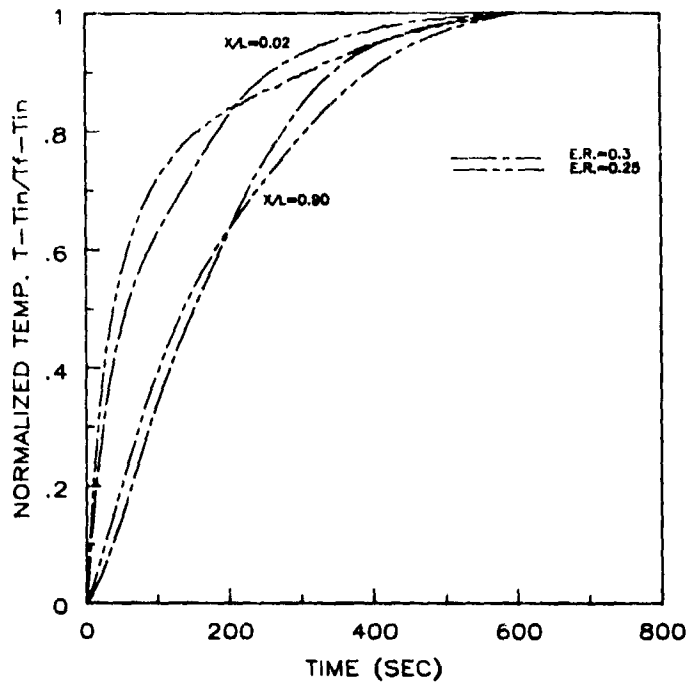

Figure 3 — Normalized temperature versus time at X/L = 0.02 and 0.90 for equivalence ratios of 0.25 and 0.30, 700°K inlet temperature and 6 m/sec reference velocity.
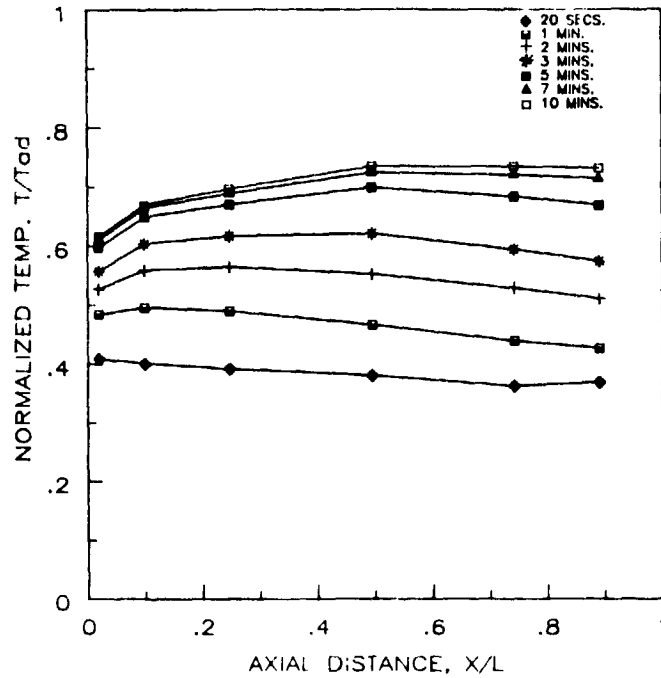
273

Figure 4 — Normalized temperature versus X/L at different times after ignition until
steady state, for 700°K inlet temperature, 0.3 equivalence ratio and 6
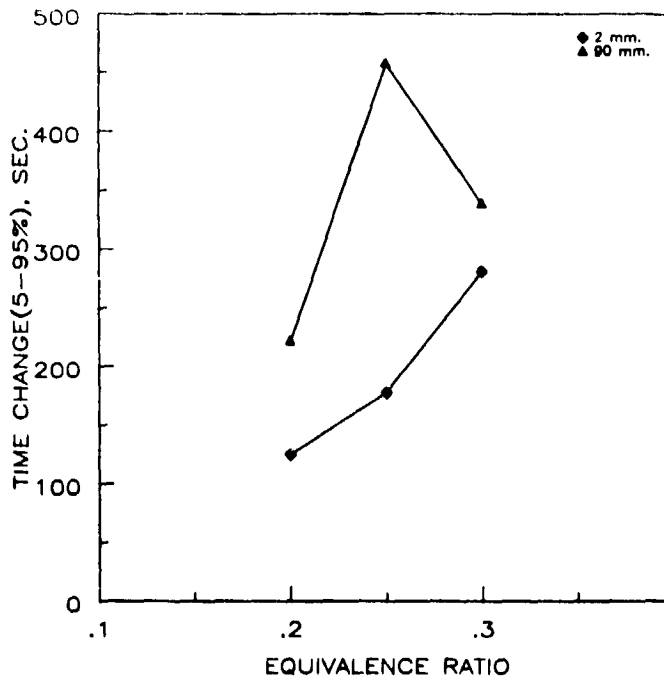m/sec reference velocity.

Figure 5 — Time required for $(T-T_{inlet})/(T_{final}-T_{inlet})$ to go from 0.05 to 0.90 ver-
sus equivalence ratio at X/L = 0.02 and 0.90 for 700°K inlet temperature
and 10 m/sec reference velocity.