Lean catalytic combustion of alkanes at short contact times ☆

C.T. Goralski Jr. and L.D. Schmidt

Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, MN 55455, USA

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The catalytic combustion of methane, ethane, propane, and *n*-butane in air over platinum coated ceramic foam monoliths at contact times ranging from 3 to 25 ms and catalyst temperatures between 800 and 1100°C has been examined for compositions leaner than the homogeneous flammability limits. The extinction limits for heterogeneous reaction for these fuels has been determined as a function of preheat temperature up to 700°C in order to simulate the performance of a catalytic combustor or incinerator with exhaust gas preheat. The effect of the superficial velocity on the extinction limits has also been examined. We observe that the extinction composition decreases with increasing preheat temperature and increases with increasing flow rate for all fuels, and the reactivity of the fuels increases with the length of the carbon chain. Methane can be made to react completely as lean as 2.3% in air, while ethane, propane, and butane can be made to react completely as lean as 0.6, 0.4 and 0.2% respectively. For butane we observe only a single steady state for preheat temperatures above 200°C and a bifurcation diagram showing the extinction behavior for this system was constructed.

Keywords: catalytic combustion; catalytic incineration; short contact time; platinum; monolith; alkane oxidation

1. Introduction

The catalytic combustion of hydrocarbons has considerable potential in applications such as catalytic combustors for power production [1], the removal of volatile organic compounds and hazardous air pollutants from exhaust air streams [2–5], and in the design of radiant heat sources [6]. Monolith catalysts [7,8] have been shown to produce high conversions and selectivities to partial oxidation products such as synthesis gas [9,10] and olefins [11,12] at millisecond contact times when operated in the fuel-rich regime.

In this work we investigate the operation of monolith catalyst systems in the fuel-lean regime where carbon dioxide and water are the only expected products for feeds of methane, ethane, propane, and *n*-butane. We use preheat to extend the catalytic ignition and extinction limits to leaner compositions and to maintain sufficiently high catalyst temperatures (800–1100°C) that reaction is complete in contact times of several milliseconds.

One of the primary advantages of catalytic combustion over traditional gas-phase combustion is the leaner compositions at which catalytic combustors can operate, resulting in fuel savings and cooler reactor temperatures. The lean flammability limit for all paraffins in stagnant ambient air is about $\phi = 0.5$ [13], where ϕ is the fuel air ratio divided by the stoichiometric fuel/air ratio for the formation of CO_2 and H_2O.

Fig. 1a shows qualitatively how the adiabatic reactor temperature should vary with percent fuel for complete fuel conversion of paraffins to carbon dioxide and water, as well as the homogeneous flammability limits for stagnant gases. As sketched in fig. 1a, the adiabatic temperature rise (ΔT_{ad}) is approximately linear with composition,

$$\Delta T_{\mathrm{ad}} \cong \frac{\Delta H_{\mathrm{R}}}{\rho C_p} C_0 , \qquad (1)$$

where C_0 is feed concentration of the limiting reactant (fuel for lean compositions), ΔH_{R} is the heat of reaction, and ρC_p is the heat capacity of the gases. The peak temperatures are > 2000°C for these fuels in air at $\phi = 1$, and homogeneous combustion chambers usually require $\phi > 0.7$ for stable operation. Operation in the lean regime allows catalytic combustors to operate at lower temperatures than homogeneous combustors whose peak temperatures usually exceed 1500°C where the formation of thermal NO_x can become significant. Additionally, lean catalytic combustors can ensure more complete conversion of fuel with no CO formation.

Conventional catalytic combustors involve noble metals deposited on high surface area alumina washcoats (up to 200 m^2/g) on extruded ceramic monoliths or porous alumina pellets, and they operate at inlet temperatures of ~ 250°C and exhaust temperatures of 300–500°C with residence times on the order of 1 s [14]. At these low operating temperatures and long contact times with small pores in the washcoat, conventional catalytic combustors are very susceptible to sintering and to catalyst poisons such as Cl and S. In contrast, the system discussed here consists of platinum coated α-Al_2O_3 monoliths without washcoats which operate at high conversions with residence times on the order of 10 ms at temperatures of 800 to 1100°C. This is a film catalyst

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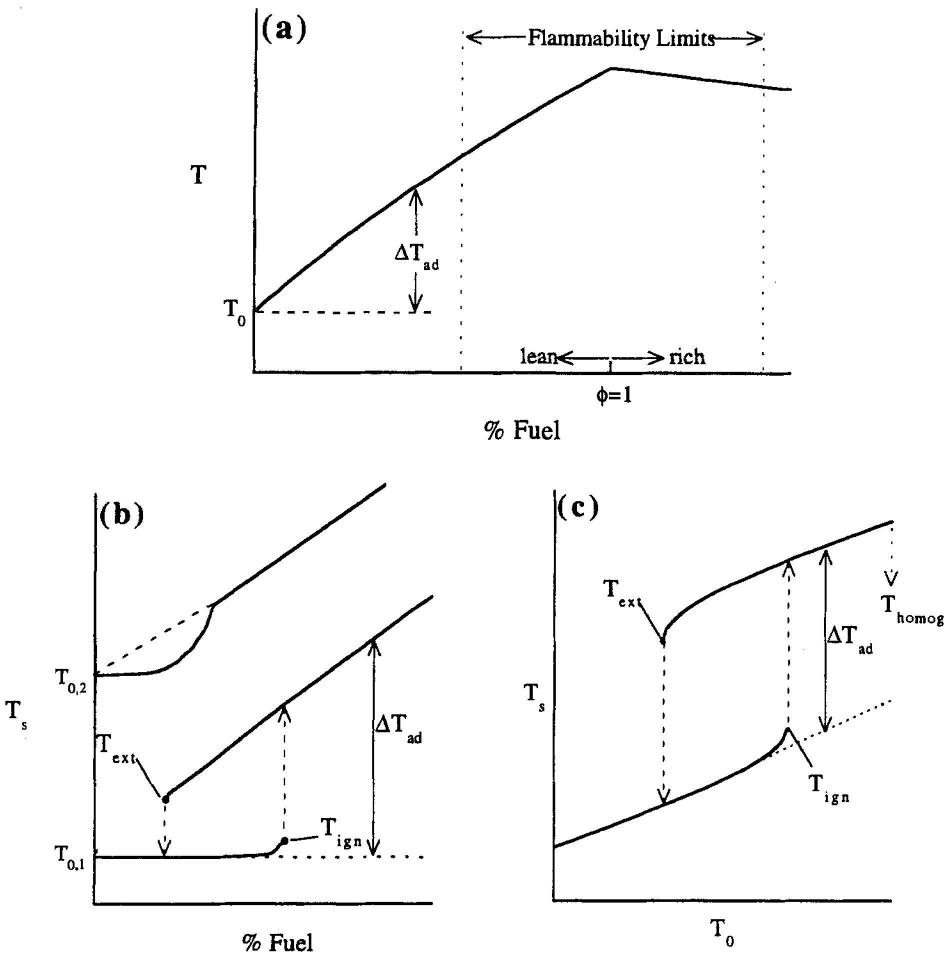

Fig. 1. Ignition/extinction behavior. (a) Adiabatic reactor temperature as a function of the inlet fuel composition. (b) Catalyst temperature as a function of inlet fuel composition showing that ignition/extinction behavior can be obtained by varying the inlet fuel composition. (c) Catalyst temperature as a function of inlet temperature showing that ingnition/extinction behavior can be obtained by varying the inlet gas temperature.

that contains no pores and has a surface area of only $\sim 1\ m^2/g$. This reactor is much less susceptible to catalyst poisons because the temperatures are higher and requires a much smaller reactor volume due to the significantly shorter residence time.

Catalytic oxidation reactions are very fast, usually mass transfer limited, and run nearly adiabaticly at short residence times. This can cause ignited and extinguished steady states in flow reactors as sketched in figs. 1b and 1c. The ignited branch operates at high temperature and conversion, while the extinguished branch operates at low temperature and low conversion. Fig. 1b shows how the surface temperature (T_s) of a catalytic oxidation reactor should vary with the inlet composition for two different preheat temperatures ($T_{0,1}$ and $T_{0,2}$), while fig. 1c shows how T_s varies with the preheat temperature (T_0). These two figures show how ignition/extinction behavior can be observed by varying the inlet composition or the inlet temperatures. The lower branch of the lower curve in each of these figures represents the extinguished branch, while the upper curve represents the ignited branch. The ignition temperature (T_{ign}) and extinction temperature (T_{ext}) are indicated on each figure. The ignited branch is the only regime of interest for most catalytic oxidation systems, and thus the extinction temperature and composition are very important to determine the operating limits of these systems.

At high preheat temperatures these reactors will exhibit only one steady state, as shown in the upper curve ($T_{0,2}$) in fig. 1b. When only one steady state exists, no hysteresis will be observed in the catalyst temperature as the composition is varied, rather the temperature will be a continuous function of composition. This behavior occurs because the rate of surface reaction is much faster at higher preheat temperatures and appreciable reaction occurs even at extremely lean compositions.

The temperature of the catalyst surface when homogeneous reaction begins (T_{homog}) is also shown in fig. 1c. The catalyst temperature will decrease once homogeneous reaction begins because the majority of the reaction takes place in the gas phase rather than on the catalyst surface.

The preheat temperature (T_0) is an important variable to study for this system because the higher the inlet temperature of the gas mixture, the leaner the catalyst will be able to operate without extinguishing.

Fig. 2 illustrates several monolith reactor configurations. The ideal reactor would consist of a monolith catalyst in a well insulated tube that would operate fuel lean. This reactor can be made to run leaner by preheating the inlet gases [15]. In practice, lean operation is most effectively accomplished by utilizing the sensible heat of the exhaust products to preheat the inlet gases, as shown in the heat exchange reactor in fig. 2b. Operation with this configuration will allow temperatures within the catalyst to rise considerably above the adiabatic temperature for an ambient inlet temperature at a given inlet composition. The amount of preheat provided to the inlet gases for this reactor configuration depends on many characteristics of the reactor, such as the length, diameter, and thermal conductivity of the wall material, making it difficult to vary the preheat temperature in a small reactor. We simulate the heat exchange reactor in the laboratory by providing preheat with an electric furnace and measuring the gas temperature just before the catalyst. This is equivalent to a reactor with recuperative heat exchange, but it is simpler to realize experimentally.

2. Experimental

The reactor consisted of a cylindrical platinum coated foam monolith catalyst 20 mm in diameter and 10 mm thick in a quartz tube reactor 40 cm long with a 21 mm inner diameter oriented vertically direction as shown in fig. 2c. The foam monoliths were 92% α-Al_2O_3, 8% SiO_2 with a void fraction of about 0.8. The monoliths were coated with 2 to 8 wt% platinum by salt impregnation followed by calcination and reduction as described previously [12]. The catalyst was placed between two inert foam monoliths which acted as radiation shields. The catalysts and heat shields had porosities of 45 pores per inch. The catalyst and radiation shields were wrapped in an Al_2O_3/SiO_2 fiber mat insulation to prevent heat loss in the radial direction as well as to prevent gases from bypassing the catalyst. The outside of the tube was insulated with an Al_2O_3/SiO_2 fiber insulation. An upstream furnace provided preheat (T_0) for the inlet gases.

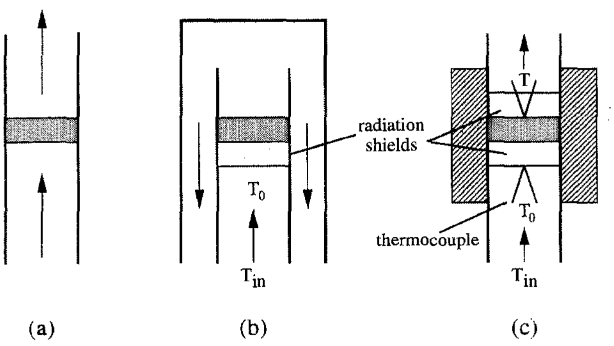

Fig. 2. Monolith reactors. (a) Typical monolith reactor. (b) Monolith reactor with exhaust gas preheat. (c) Laboratory reactor.

The temperature T of the catalyst was measured on the downstream face of the catalyst before the downstream heat shield with a bare Pt/Pt–13% Rh thermocouple with an estimated accuracy of approximately ±20°C. The preheat temperature T_0 of the gas mixture was measured with a sheathed thermocouple (otherwise the thermocouple becomes catalytic) with the junction ~ 1 cm before the upstream heat shield. The preheat temperature was controlled using a temperature controller. The gases were premixed with flow rates measured using mass flow controllers, and flow rates are reported as standard liters per minute (SLPM). Residence time of the gases and velocities in the catalyst were calculated by accounting for expansion. For example, 5 SLPM at 1000°C corresponds to $\tau \approx 5 \times 10^{-3}$ s and $u \approx 2$ m/s within the catalyst. All experiments were run at pressures of 1 atm. The product gases were exhausted into a fume hood. Compositions of the exhaust gases were measured using a Hewlett Packard 5890 gas chromatograph with a thermal conductivity detector. This confirmed that all hydrocarbons were completely consumed ($>$ 99.5% conversion) when the catalyst was ignited and that CO_2 and H_2O were the only products observed for all compositions examined.

The experiments were started by fixing the gas flow rates and compositions and then setting the desired preheat temperature. Once the inlet temperature of the gases stabilized, the catalyst was ignited by holding a burner near the catalyst until the catalyst was visibly ignited. The burner was then removed, the catalyst zone insulated, and the catalyst ran autothermally. Methane has an ignition temperature of 600°C on Pt, making ignition difficult. However, ammonia has an ignition temperature of 200°C on Pt [12,18], and therefore in order to ignite methane mixtures, ~20% ammonia was typically added to the gas mixture. Once the catalyst was ignited, the ammonia was shut off and the catalyst ran with methane as the only fuel. Ethane, propane and butane were usually ignited without ammonia because these fuels have lower ignition temperatures on Pt than methane [17].

Experiments were run with catalysts that had weight loadings of 2, 5 and 8 wt%. The results for each of the catalyst loadings were similar, and all results presented here are for a catalyst loading of 5 wt%.

3. Results

3.1. Preheat

In these experiments the catalyst was ignited at a fixed preheat temperature and then the composition of the inlet mixture was slowly made leaner until the catalyst temperature was observed to drop rapidly by over 100°C (fig. 1b). This composition and catalyst temperature were then recorded as the extinction composition and temperature for the given preheat temperature.

As expected, the catalyst would remain ignited at lower compositions when the inlet temperature of the gases was higher. The extinction temperature of the catalyst varied strongly with the preheat temperature for methane and ethane. However, propane showed a relatively constant extinction temperature. Butane showed more complicated behavior than methane, ethane, and propane, exhibiting no hysteresis at higher preheat temperatures.

3.1.1. Methane

The effect of preheat on the extinction composition was studied at a flow rate of 5 SLPM, corresponding to a residence time of ~5 ms, and data are plotted in fig. 3a. The plot also shows the lean homogeneous ignition limit. This lean limit corresponds to ignition in stagnant air, and the limit for this flow system will be shifted considerably to the right because the flame speed must exceed the superficial velocity of the gases in the reactor in order for the flame to propogate. As can be seen from the plot, the "window" of operation between the homogeneous ignition limit and the heterogeneous extinction limit is at least 0.6% composition and remains largely constant with increasing preheat temperature. In fact, experiments showed that operation up to 3% richer in composition than the stagnant limit is possible with no obvious flames. Gas chromatograph analysis showed complete conversion of methane for all regions where the catalyst was ignited.

Fig. 3b shows a plot of the catalyst extinction temperature as a function of the preheat temperature. The catalyst extinction temperature was found to decrease strongly with the preheat temperature.

3.1.2. Ethane

Fig. 4a shows a plot of the preheat temperature versus the composition where extinction occurred for ethane in air at 5 SLPM. For ethane, the region between heterogeneous extinction and the calculated static homogeneous ignition limit was only ~0.3% at low preheat and almost 0.7% at high preheat. However, ethane was routinely operated up to 2% over the static limit without any visible homogeneous reaction. Gas chromatograph analysis showed complete conversion of ethane for all regions where the catalyst was ignited.

Fig. 4b shows the variation in the catalyst extinction temperature with preheat temperature for ethane. As with methane, the surface extinction temperature dropped by almost 300°C as the preheat was increased from 200 to 650°C. It can be observed that the ethane system extinguishes about 100°C cooler than the methane system at a given preheat temperature.

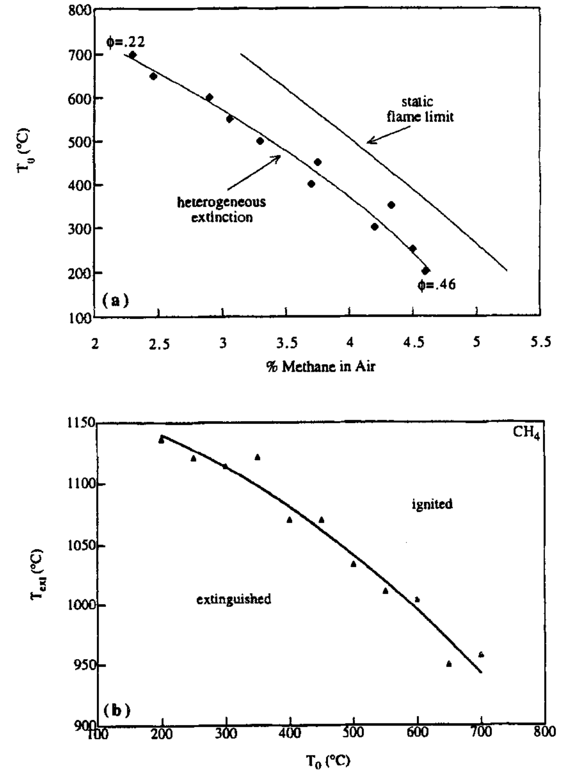

Fig. 3. Methane extinction. (a) Extinction composition as a function of preheat temperature. (b) Catalyst extinction temperature as a function of preheat temperature.

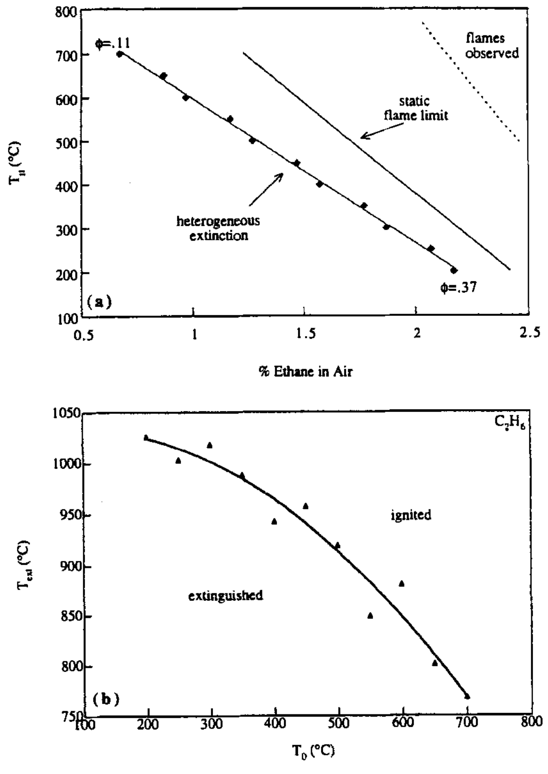

Fig. 4. Ethane extinction. (a) Extinction composition as a function of preheat temperature. (b) Catalyst extinction temperature as a function of preheat temperature.

3.1.3. Propane

Fig. 5a shows a plot of preheat temperature versus the extinction composition for propane at 5 SLPM. For propane, it was observed that the window of operation between the extinction limits and the static lower ignition limits was about 0.5%, and remained approximately constant with temperature. Propane could be run up to 1.5% richer than the static ignition limit without visible homogeneous reaction. Gas chromatograph analysis showed complete conversion of propane for all regions where the catalyst was ignited.

The effect of preheat on the extinction temperature of the catalyst for propane is shown in fig. 5b. The behavior observed here was significantly different than the behavior observed for methane and ethane because the catalyst extinction temperature was almost independent of the preheat temperature. Propane also ran up to 350°C cooler than methane and ethane at low preheat.

3.1.4. Butane

Butane was found to be significantly more reactive than the other fuels examined. At preheat temperatures above ~200°C at a flow rate of 5 SLPM, no clear extinction temperature was observed, indicating that only one steady state existed for these inlet conditions. No discernible extinction temperature was seen until the preheat temperature was lowered to 180°C. Fig. 6 shows a plot of catalyst temperature versus the fuel composition for butane oxidation at various preheat temperatures. It is clear that above 200°C of preheat there was a continuum of temperatures with varying composition with no ignition/extinction hysteresis. Gas chromatograph analysis showed complete conversion of butane for catalyst temperatures above ~500°C but incomplete conversion at catalyst temperatures cooler than this. This figure shows only the extinction branch of the bifurcation diagram sketched in fig. 1b. The behavior below 200°C displaying two steady states corresponds to the lower curve in fig. 1b ($T_{0,1}$), while the data above 200°C is indicative of the single steady state behavior shown in the upper curve ($T_{0,2}$).

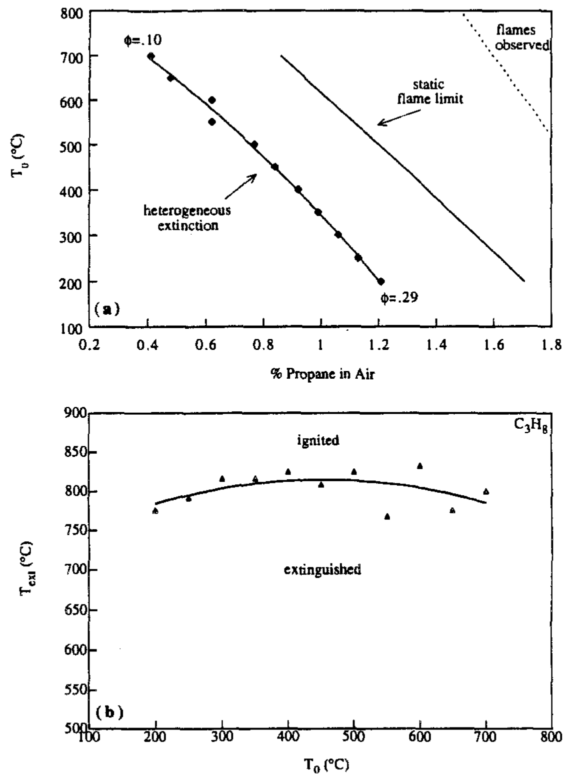

Fig. 5. Propane extinction. (a) Extinction composition as a function of preheat temperature. (b) Catalyst extinction temperature as a function of preheat temperature.

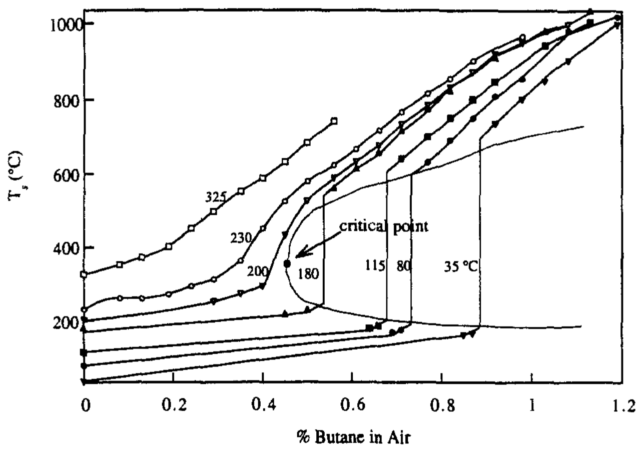

Fig. 6. Butane extinction. Catalyst temperature as a function of fuel composition and preheat temperature.

For methane and ethane, two steady states were observed for all preheat temperatures examined. Propane showed two steady states for all results shown in fig. 5a. However, at a preheat temperature of 750°C, propane exhibited only one steady state. Higher preheat temperatures were not examined because of limitations of the experimental system.

3.2. Comparison of four fuels

Fig. 7 is a comparison of the preheat temperature versus the heterogeneous extinction compositions in terms of ϕ for methane, ethane, propane, and butane which shows that the heterogeneous extinction limit varies strongly with the fuel. Butane is significantly more reactive than the other three fuels, showing no extinction point at preheat temperatures above 200°C. For homogeneous combustion, the static lean ignition limit is about $\phi = 0.50$ for all paraffins at ambient conditions, and varies approximately linearly with temperature [13]. The difference in the reactivities for the various fuels that is observed for heterogeneous combustion suggests that the mechanisms that control heterogeneous combustion

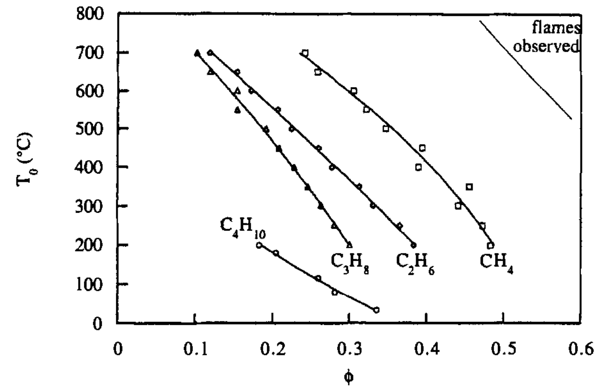

Fig. 7. Comparison of four fuels examined. Extinction compositions in terms of ϕ as a function of the preheat temperature.

are significantly different than for homogeneous combustion.

3.3. *Effect of flow rate*

The effect of the flow rate on the extinction limits for ethane oxidation was also examined, and fig. 8a shows a plot of the catalyst temperature as a function of ethane composition for flow rates between 1 and 7 SLPM. The calculated adiabatic temperature as a function of composition is also plotted on this graph. This figure shows that the extinction composition (the break in the curves) varies significantly with flow rate. The plot also shows that as the flow rate was increased, the surface temperature moved closer to the calculated adiabatic temperature. This behavior is consistent with radiation as the primary mechanism of heat loss. Fig. 8b shows a plot of the extinction composition versus flow rate taken from the compositions where the temperature curve breaks sharply downward in fig. 8a. This plot shows a nearly linear increase in the extinction composition with flow rate. Below 2 SLPM there is sufficient heat loss that only one steady state exists. We observed qualitatively similar results for methane, propane, and butane.

The measured catalyst temperatures for flows of 5 SLPM showed that the reactor ran between 80 and 90% of the calculated adiabatic temperature shown in fig. 1 with the difference due mostly to radiation losses. A plot of the log of the catalyst temperature versus the log of the flow rate showed a straight line with a slope of 0.27. Since radiation is proportional to T^4, if all the heat loss were due to radiation the slope should be 1/4. These results are consistent with radiation as the primary mechanism of heat loss.

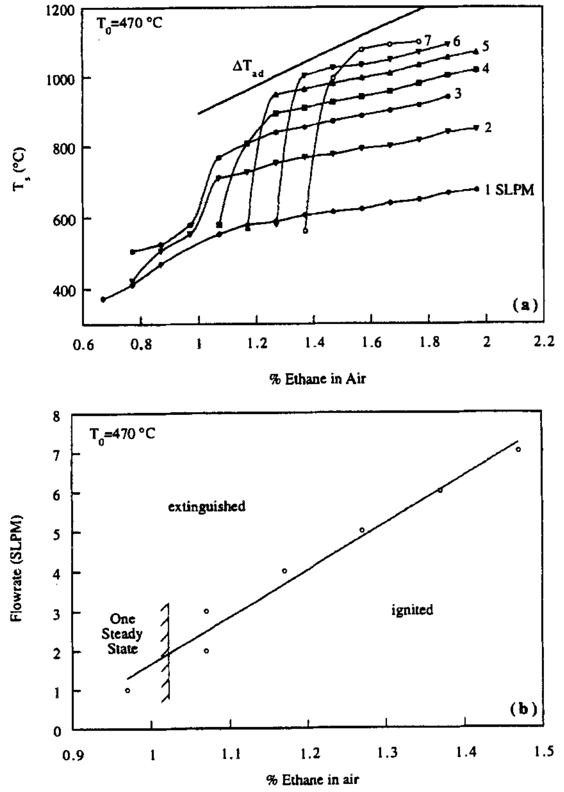

Fig. 8. Effect of flow rate on extinction composition. (a) Catalyst temperature as a function of fuel composition and flow rate for ethane at a preheat temperature of 470°C. (b) Extinction composition as a function of flow rate for ethane in air at a preheat temperature of 470°C.

3.4. *Homogeneous ignition*

Homogeneous ignition was observed in the reactor in the upstream and downstream directions when the fuel composition was moved far enough into the regime of homogeneous flammability. Upstream flames typically started in the preheat zone near the upstream radiation shield and would propagate back to the mixing point and extinguish. Upstream flames were accompanied by a decrease in the catalyst temperature because fuel was consumed before the catalyst. Downstream flames typically existed as standing flames above the downstream heat shield. Obviously, downstream flames can only exist if fuel breakthrough occurs, and downstream flames were only observed when the catalyst had completely extinguished. Downstream flames could then only be extinguished by shutting off the fuel supply to the reactor or by reigniting the catalyst with an external heat source.

As expected, methane was found to be the most resistant to homogeneous ignition. Heterogeneous methane reaction could be sustained without flames at compositions up to $\phi = 0.9$ with no preheat, though some catalyst loss and degradation of the monolith support were seen at these conditions because of the high temperature (~1800°C) associated with these compositions. Ethane and propane were also quite stable, but at about $\phi = 0.7$ upstream flames were observed at preheat temperatures above 500°C. Butane was most reactive, with upstream flames observed for $\phi = 0.5$ at preheat temperatures over 500°C. These compositions are all significantly richer than the stagnant flammability limits for the given preheat temperatures, indicating that catalytic operation well into the stagnant flammability limits without flames is possible.

The upstream alumina heat shield was occasionally observed to ignite at very high catalyst temperatures. This behavior was typically observed at preheat temperatures above 500°C at low flow rates. This only occurred when the catalyst was near the extinction composition and the heat shield and catalyst then extinguished within several minutes. At these operating conditions, the majority of the reaction takes place on the front of the catalyst, leading to high surface temperatures near the upstream heat shield. Heat conduction upstream could then heat the radiation shield enough that the bare alumina exhibits catalytic activity.

3.5. Other materials

Different catalyst support materials and catalysts were also examined qualitativly. Platinum coated cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) foam monoliths were examined for methane oxidation, although these monoliths melted when ignited for methane compositions near $\phi = 0.5$. At this composition, the calculated gas phase adiabatic temperature is ~1200°C, which is below the melting point of cordierite, 1360°C. The melting of the cordierite catalyst was likely a result of local surface temperatures that exceeded the gas phase adiabatic temperature.

Rhodium and palladium were also examined as catalysts. Rhodium was found to have no catalytic activity in the fuel lean regime (no ignition) which result is supported by other ignition extinction research in this laboratory [17]. This is likely because the rhodium exists as Rh_2O_3 which exhibits little catalytic ativity in the oxidizing atmospheres examined here. Palladium seems promising as an oxidation catalyst, and initial results indicate that palladium operates even leaner than platinum. In this case, PdO is actually significantly more active than Pd, requiring that surface temperatures be maintained below ~950°C where the oxide decomposes [19]. We are continuing further examination of palladium and other metals, as well as other support materials in this reactor.

4. Discussion

The majority of the literature dealing with the catalytic combustion of alkanes deals with examination of the conversion of fuel over a catalyst bed of noble metals on porous monoliths or spheres as a function of the inlet gas temperature at relatively low temperatures for residence times of ~ 1 s [4,16]. Because of the cool temperatures at which most catalytic combustors are operated (< 500°C) the reactions are typically limited by the surface reaction rates or mass transfer in the pores. Operation at high conversion in these catalysts is a problem because the high surface temperatures associated with ignition causes the dispersed catalyst particles and γ-Al_2O_3 to sinter, thus blocking pores and causing catalyst deactivation.

Experiments here differ significantly from those experiments because the conversion in all cases is complete with temperatures approaching adiabatic with no dilution. This is because we use a film catalyst where the reaction rate is limited by bulk mass transfer to the surface of the catalyst, allowing us to operate at very short contact times. We are primarily concerned with the extinction limits and the onset of homogeneous reaction because these are the lower and upper operating limits of such a reactor.

4.1. Extinction compositions

The data for the extinction limits of methane, ethane, propane, and butane show that the reactivity of the fuel increases with carbon chain length, as expected. This is most clearly seen in fig. 7 which shows that propane will remain ignited at compositions further from stoichiometric compared to methane and ethane. Most ignition/extinction mechanisms for paraffins on platinum suggest that before ignition, the surface of the catalyst is covered with mostly atomic oxygen, and the removal of the oxygen is the essential step in ignition [16,17], while after ignition the surfaces are nearly clean. If it is assumed that the slow step on the ignited catalyst is the fuel molecule dissociation and adsorption on the surface, then fuels that have weaker C–H bonds, and therefore more stable radical species, would be more reactive than fuels that have stronger C–H bonds.

The butane data presented in fig. 6 indicates that at preheat temperatures over 200°C only one steady state exists for lean compositions because the higher reactivity of butane allows significant conversion without ignition, even at extremely lean compositions. For methane, ethane, and propane, the conversion of fuel on the extinguished branches is extremely low, typically less than 5% even at a preheat temperature 650°C. Because of the low conversion, little heat is generated, and the catalyst remains cool. Butane, on the other hand, shows nearly 40% conversion and a catalyst temperature of over 400°C at just 0.2% fuel for a preheat temperature of 325°C. At these conditions, the catalyst is not completely ignited, and the reaction rate is controlled by the rate of butane reaction on the surface. However, the heat liberated by the reaction is not sufficient to accelerate the rate enough to provide complete conversion of the fuel. The catalyst ignites only when the composition is increased such that enough heat is generated to accelerate the surface reaction so that mass transfer is the limiting step in reaction. At lower preheat temperatures, the reaction rate on the extinguished branch is slow as for the other fuels, and butane shows similar extinction behavior as methane, ethane and propane.

Fig. 6 shows an envelope where two steady states will exist for butane combustion over monolith catalysts. When operating outside this envelope, only one steady state will exist. The figure also shows a critical point on the envelope at a preheat temperature of ~195°C and composition of ~0.45%. This critical point indicates that at preheat temperatures over 195°C or compositions leaner than 0.45%, only one steady state will exist.

4.2. Homogeneous reaction

Homogeneous combustion reactions are free radical chain reactions with chain branching. Clearly, at the high temperatures that exist within the catalyst structure, most of the fuel mixtures discussed here would be flammable at the feed composition and temperature. However, because reaction removes fuel quickly and the pores of the catalyst are smaller than the quenching distance for a flame, we believe that there is insignificant homogeneous reaction within the ignited catalyst. This leaves flames in the regions of the reactor upstream and downstream of the reactor as the only parts of the reactor where homogeneous reaction is likely to take place. Flames downstream of the catalyst were only observed once the catalyst was extinguished. This was typically initiated by upstream flames that consumed much of the fuel in the inlet mixture and caused the catalyst to cool and extinguish. In these experiments, operation well into the flammability limits has been demonstrated without significant problem with upstream flames. These limits could be pushed even further by packing the inlet section of the reactor with an inert material to suppress upstream flames.

4.3. Lean catalytic incineration

The data in these experiments provide much of the information necessary to begin the design of a catalytic incinerator to destroy volatile organic compounds in dilute concentrations in an air stream. The reactor described here would be ideal for this application because the high temperatures of operation allow for very efficient destruction of the organic pollutant and would require minimal additional fuel because of the ability of the reactor to operate lean. For an incinerator, the exhaust gases would be used to preheat the inlet gases, further lowering the fuel requirement. An 1 ft^2 catalyst in an incinerator of this type could easily handle a flow rate of 4000 CFH of contaminated air under conditions of these experiments. The most obvious choice of the additional fuel would be either methane (the main component in natural gas) or propane (liquified petroleum gas) because of these fuels are readily available at low cost. From the results presented here, propane seems to be more attractive because of the leaner operation and lower catalyst extinction temperature at low preheat temperatures, making control of the inlet gas merely a matter of maintaining a constant catalyst temperature of ~900°C by controlling the fuel flow rate. Natural gas is equally viable as a fuel, although the control of the catalyst temperature would be more complicated because of the preheat temperature dependance of the catalyst extinction temperature.

4.4. Ignition

The ignition characteristics of this system were not systematically examined. This is because ignition is of little practical interest in the design or operation of a catalytic combustor of this type. The catalyst would most likely be ignited with a homogeneous flame until the temperature of the catalyst becomes hot enough to support the hetergeneous reaction, at which time the flame would be extinguished by lowering the fuel composition.

5. Conclusions

Catalytic combustion over monolith catalysts at short contact times allows for complete conversion of lean fuel mixtures at high temperature in reactors that are significantly smaller than conventional catatytic combustors. These systems are very stable at high temperatures and are inherently poison resistant. These systems can run from compositions as lean as 10% of the stoichiometric composition for formation of carbon dioxide and water, up to compositions that are nearly 90% of the stoichiometric composition without the presence of homogeneous flames. This type of reactor could be used for many potential applications including catalytic incinerators, power sources, or radiant heat sources.

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