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Comparative study of lean and rich filtration combustion of methane/air, ethane/air, and propane/air mixtures

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Abstract: - This work comparatively analyzes lean, rich and ultrarich combustion of methane (CH_4)/air, ethane (C_2H_6)/air and propane (C_3H_8)/air mixtures inside inert porous media. Temperature, velocity, and chemical products of the combustion waves were studied experimentally in the range of equivalence ratios (ϕ) from 0.2 to 2.5, at a single filtration velocity of 12 cm/s. Low-velocity regime (LVR) of filtration gas combustion was observed for (CH_4)/air and (C_3H_8)/air mixtures for the whole range of the studied equivalence ratios . Transition to high-velocity regime (HVR) occurs in the range $0.9 < \phi < 1.15$ for (C_2H_6)/air mixtures. Methane (CH_4)/air mixtures have essentially higher combustion temperatures than corresponding (C_2H_6)/air and (C_3H_8)/air mixtures. The products of methane, ethane and propane partial oxidation (H_2, CO, and C2 hydrocarbons) are dominant for ultrarich superadiabatic combustion where up to 60%, 70%, 55% of fuel, respectively, is converted to CO and H_2. Ethane conversion to hydrogen was significantly higher than for methane and propane.

Keywords: - Porous media burner; Hydrogen; Combustion; Partial oxidation

1. Introduction

It has been well recognized by a number of researchers that there are practical benefits of combustion within a porous media, due to its clean and highly efficient characteristics. During the past 20 years a variety of works has been published that describe gaseous fuel combustion in porous media [1-3]. Within this context, stationary and transient systems are the two major design approaches commonly employed in porous combustion. The first approach is widely used in radiant burners and surface combustor-heaters where the combustion zone is stabilized within the finite element of the porous matrix. The second (transient) approach involves a traveling wave where the unsteady combustion zone freely propagates as a wave either downstream or upstream within the packed bed.

As a result of intensive interfacial heat transfer between the gas and the solid, observed combustion temperatures can be significantly different from the adiabatic predictions and are controlled mainly by the reaction chemistry and heat transfer mechanism. These conditions correspond to the LVR of filtration gas combustion, according to classification given by Babkin [1]. LVR, HVR, sound velocity regime (SVR) and other regimes are distinguished in literature [1,4] by the velocity of the combustion front relative to solid phase and peculiarities of the heat and mass transfer in the system. Most experimental and modeling studies have been performed in the LVR where flame propagation speeds are on the order of 10^{-4} m/s. On the other hand, when the heat exchange is rather weak, LVR converts into HVR where characteristic flame speed is ~ 10 m/s. Compared to LVR, the more important factor in HVR is not thermal but aerodynamic interphase interaction and its consequences. Thermal interaction has actually no direct effect on the chemical reaction except for the most rapid pulsations. Therefore, in the zone of chemical reaction, there is no recuperation of heat that is typical of LVR.

The applications of porous combustion developed over the last few years include porous radiant burners, porous media combustor-heaters, porous burners for pollution control, and reverse-flow filtration burners (RFB). In RFB technique, a confined wave is allowed to propagate along the solid. It is controlled by periodically reversing of the reactants flow. Currently, several gaseous fuels have been tested using this method [5]. A disadvantage of this technique compared to a stationary flame system may be its relatively large size and high number of thermal cycles that the porous media is subject to. Other modern applications of transient porous burners include combustion of low-calorific fuels [6], VOC emissions control with the possibility of heat recovery [7], and fuel reforming in the ultrarich superadiabatic flames [5,8].

At rich conditions, the superadiabatic combustion temperatures create a situation in which partial oxidation and/or thermal cracking of hydrocarbons can occur [2]. Multiple studies have shown that the conversion of methane to hydrogen occurs in two stages: initially through partial oxidation and then by steam reforming. Partial oxidation of methane produces hydrogen, and the heat generated in the partial oxidation can supply the energy for steam reforming, generating more hydrogen [5]. The vast majority of work in fuel reforming through filtration combustion has been done using the packed bed design. The Kennedy's group at the University of Illinois at Chicago has conducted extensive research in transient filtration combustion and the production of hydrogen in packed bed reactors [9]. Recent work by the group utilizes a reciprocal flow burner (RFB) which periodically reverses the flow direction to restrict the propagating wave front to a fixed volume of aluminum oxide beads [5]. Further studies were done by the group for hydrogen production in ultra-rich filtration combustion of methane and hydrogen sulfide [8,10] and filtration combustion in oxygen enriched and oxygen depleted environments [11]. The particularly interesting area of ultrarich filtration combustion of complex fuels remains unexplored.

In this work, the results of a comparative study of filtration combustion from lean to rich methane/air, ethane/air and propane/air mixtures are presented. Temperature profiles and combustion wave velocities were measured experimentally, as were the chemical product compositions. Particular interest is to use the reactor to convert a rich fuel/air mixtures to a syngas consisting of H_2, CO, CO_2, and various other compounds.

2. Experimental apparatus

Experiments on filtration combustion wave propagation were conducted using the setup schematically shown in Figure 1. The apparatus consisted of porous heating burner, a fuel and air supply system, a temperature measurement system, and gas emission analyzers. Burner is a quartz tube with internal diameter of 38 mm and wall thickness of 2 mm. The combustible mixture was prepared by a continuous flow method where the concentrations of fuel and air were monitored using a set of MKS mass flow controllers (Model 1179A, MKS Inc.). The packed bed of 5.6 mm solid Al_2O_3 spheres was used as a porous body, it had a porosity of approximately 40%. The inner surface of the combustion tube was covered with a 2 mm layer of Fiberfrax insulation. To prevent heat losses and achieve quasi-uniform temperature profiles, additional 30 mm thick high-temperature insulation was applied on the external diameter of the reactor.

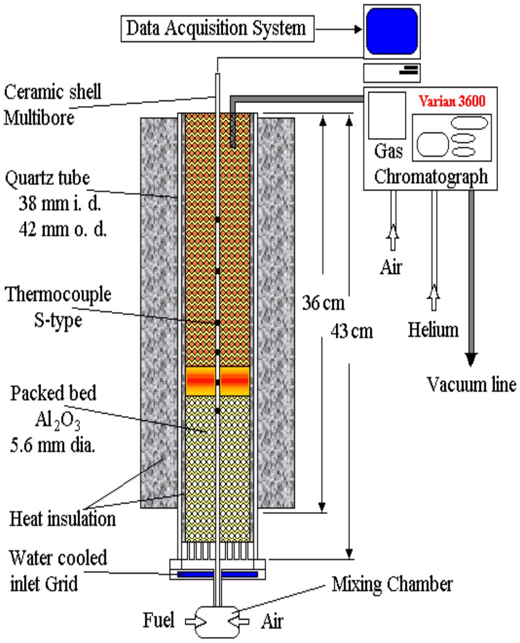

Fig. 1. Schematic of the experimental setup.

Combustion mixtures were introduced in the reactor through the distribution grid at the reactor bottom via a mixing chamber to ensure uniform gas composition. During the experiments, the combustion wave was initiated at the reactor outlet. Then upstream propagation was recorded. As the wave reached the reactor bottom, the experimental conditions were adjusted to required settings and downstream propagating wave was recorded on the reverse run. Methane/air, ethane/air, and propane/air mixtures were used in the present study.

Running axially in the centre of the quartz tube was a ceramic shell of 0.32 cm outer diameter and 56 cm long. The shell contained six holes of 0.08 cm diameter each holding an S-type (platinum/rhodium) thermocouple. Temperature measurements from these thermocouples were transferred to a PC where they were digitized using an Analog Instrument's RTI-800 board. The thermocouple junctions were equally spaced along the length of the shell, with the bottom one being 13 cm away from the gas inlet. The completely covered by ceramic material junctions recorded temperatures very close to the temperatures of the solid phase. Axial positioning of thermocouples allows minimal disturbances of gas flow and heat fluxes in the reaction zone. The experimental error in the temperature measurements was estimated as 50 K; the error in the wave velocity measurement was ~10%.

Combustion products were used at the low-temperature exit zone of the reactor and then directed for chemical analysis to a gas chromatograph (Varian 3600 GC). A ceramic tube (alumina) connected to a vacuum line was used as a sample probe. To avoid exterior air vortices, the probe was inserted 2 cm into the alumina bed. The accuracy of chemical sampling was close to 10%.

The exit of the reactor is open to atmosphere while the bottom can be plugged by either a Teflon plug or a metal one with an inner cooling system.

3. Results and Discussion

Data were collected at filtration velocity of 12 cm/s and equivalence ratios ranging from 0.2 to 2.5. The experiments were identical for three fuel/air mixtures in procedure and apparatus. Solid phase temperatures as well as product compositions were measured and the propagation rate was obtained from measured evolution of temperature profiles.

3.1 Combustion temperature and propagation rate

3.1.1 Methane

Downstream, upstream, and standing waves were observed, depending on experimental parameters, mainly the equivalence ratio. Fig. 2 displays combustion waves velocities, obtained from experimental temperature profiles; the calculated adiabatic temperatures are provided for reference. Downstream superadiabatic wave propagation of lean mixtures was observed experimentally for the region of $\phi < 0.45$. The velocity of the wave decreases with an increase of the methane concentration, approaching zero at $\phi \approx 0.45$. Under these conditions a standing combustion wave is formed. Upstream underadiabatic regime of propagation is observed for the range of equivalence ratios from 0.45 to 1.45. LVR is observed in all the range of equivalence ratios and close to 1.0 a maximum absolute value of velocity is observed. Transition to the rich combustion mixtures results in a decrease of the wave velocity; and a rich standing wave is observed for $\phi \approx 1.5$. The downstream superadiabatic propagation occurs again in the ultrarich region, where the speed increases with methane content. It was found experimentally, that the maximum combustion temperature remains constant ($T_c \approx 1370$ K) throughout the lean region and is practically independent of the methane content. However, a temperature increase (approximately 260 K) is observed with transition from ultralean to ultrarich mixtures (Fig. 2). Significantly higher temperatures ($T_c \approx 1630$ K) characterize combustion at $\phi > 1.4$. This rise of the combustion temperatures is attributed to the changes of combustion chemistry, because all other governing parameters such as flow rates, porous body properties, and heat contents are similar.

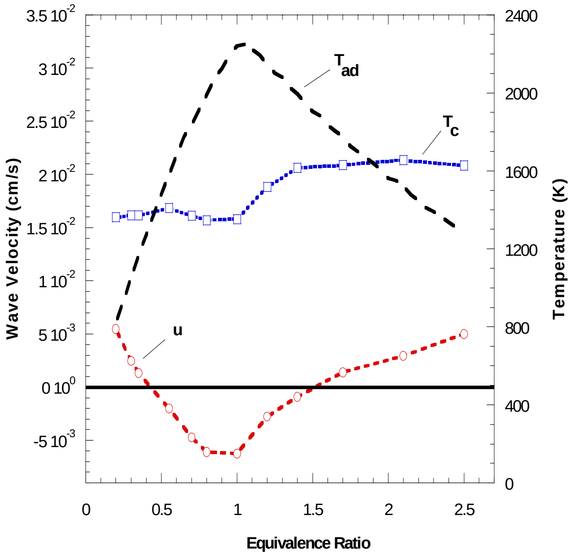

Fig. 2. Experimental data for temperatures (T_c) and velocities (u) vs. equivalence ratio for methane-air filtration combustion waves. The calculated adiabatic temperature (T_{ad}) is also plotted for reference.

3.1.2 Ethane

In Fig. 3 the experimental results for measured combustion temperatures and wave velocities are displayed for ethane/air mixtures. The experimental results, starting at $\phi = 0.2$, which is the lower limit for stable combustion, shows downstream superadiabatic wave propagation for the region of $\phi < 0.4$. The velocity of the wave decreases with an increase of the ethane concentration. A standing combustion wave is formed at $\phi \approx 0.4$. With further increase of the ethane content, the regime of propagation changes to upstream. This regime is observed for the range of equivalence ratios from 0.4 to 2.0. Transition to HVR is observed for the range of equivalence ratios $0.9 < \phi < 1.15$. Decrease of the wave velocity is observed with the change to rich combustion mixtures, and a rich standing wave is observed at $\phi \approx 2.0$. The downstream propagation occurs again in the ultrarich region, where the speed increases slowly with ethane content. Throughout the lean mixtures the maximum combustion temperature is $T_c \approx 1338$ K at $\phi \approx 0.55$. Due to the transition to HVR this temperature decreases to 1023 K at $\phi \approx 1.1$. With the change from rich to ultra-rich mixtures the temperatures increased up to $T_c \approx 1470$ K at $\phi \approx 1.7$ and then is practically independent of the ethane content. Finally, downstream superadiabatic combustion is observed for $\phi > 2.0$.

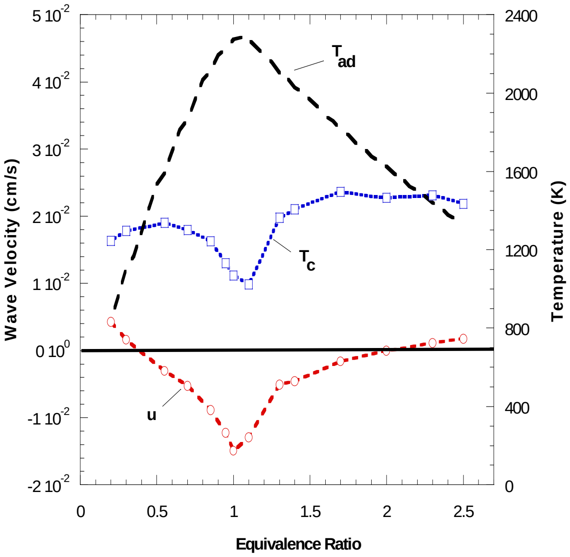

Fig. 3. Experimental data for temperatures (T_c) and velocities (u) vs. equivalence ratio for ethane-air filtration combustion waves. The calculated adiabatic temperature (T_{ad}) is also plotted for reference.

3.1.3 Propane

Downstream superadiabatic wave propagation for the region of $\phi < 0.45$ is observed in Fig. 4. Under $\phi \approx 0.45$ a standing combustion wave is formed. Upstream underadiabatic regime of propagation is observed for the range of equivalence ratios from 0.45 to 2.0. A maximum absolute value of velocity (~0.01 m/s) is observed at $\phi = 0.9$. Transition to the rich combustion mixtures results in a decrease of the wave velocity; and a rich standing wave is observed for $\phi \approx 2.0$. The downstream propagation occurs again in the ultrarich region, where the speed increases with propane content. Throughout the lean mixtures the maximum combustion temperature is $T_c \approx 1356$ K at $\phi \approx 0.5$. This temperature decreases to 1276 K at $\phi \approx 0.9$. With the change from lean to ultrarich mixtures the temperatures increases again reaching $T_c \approx 1530$ K at $\phi \approx 1.6$ and then remains practically independent of the propane content. Finally, downstream superadiabatic combustion is observed for $\phi > 2.0$.

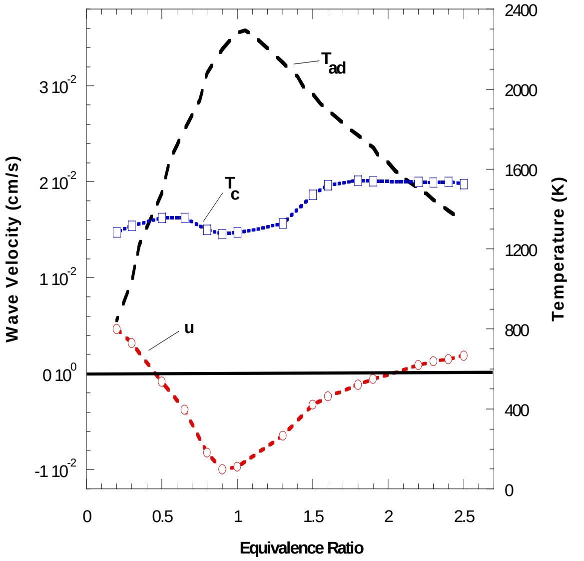

Fig. 4. Experimental data for temperatures (T_c) and velocities (u) vs. equivalence ratio for propane-air filtration combustion waves. The calculated adiabatic temperature (T_{ad}) is also plotted for reference.

3.2 Combustion products

3.2.1 Methane

Starting from equivalence ratios above 1.0, complete combustion cannot be achieved due to the low oxygen content of the mixture. Fig 5 shows that the mole fractions of H_2, CO, and CH_4 are increased with increasing equivalence ratio while the mole fraction of CO_2 drops. Significant concentrations of C_2 hydrocarbons appear in the exhaust gases. As shown in

Fig. 5, CH_4 is only generated at temperatures greater than 1600 K. At $\phi=2.1$ the largest mole fraction of hydrocarbons detected was methane at 0.6% with minute amounts of other hydrocarbons. Fig. 6 shows that the degree of methane conversion to hydrogen could be as high as 40% for highest equivalence ratios studied.

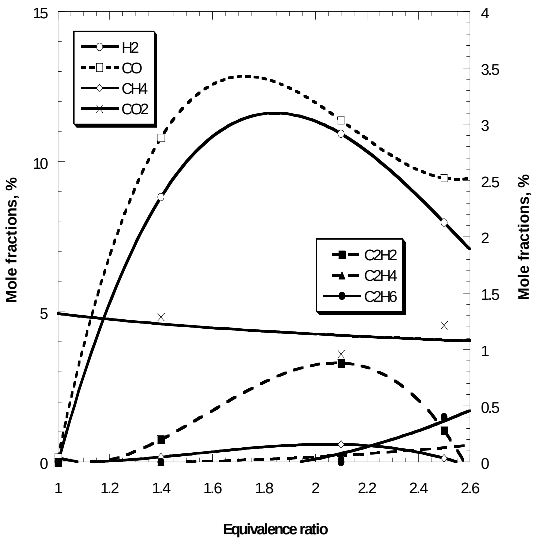

Fig. 5. Measured composition of chemical products as a function of equivalence ratio for rich and ultra-rich methane-air mixtures.

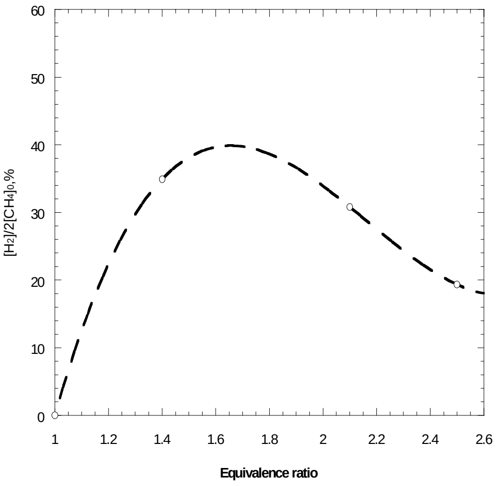

Fig. 6. Degree of methane conversion to hydrogen for rich and ultra-rich waves.

3.2.2 Ethane

The major and minor combustion products are presented in Fig. 7. The experimental results show increasing H_2 and CO and decreasing CO_2 with increase of the equivalence ratio. Significant concentrations of C_2 hydrocarbons appear in the exhaust gases with increasing of equivalence ratio. CH_4 mole fraction increased from 0.17% to over 1.73%, C_2H_2 from 0.36% to over 1.96% and C_2H_4 from 0.06% to over 040%, at φ of 1.3 and ϕ of 2.3, respectively. At ϕ =1.9, the major mole fraction of C_2H_6 is detected at 0.05%. Hydrogen conversion peaked at 40% in the rich range of equivalence ratios (Fig. 8).

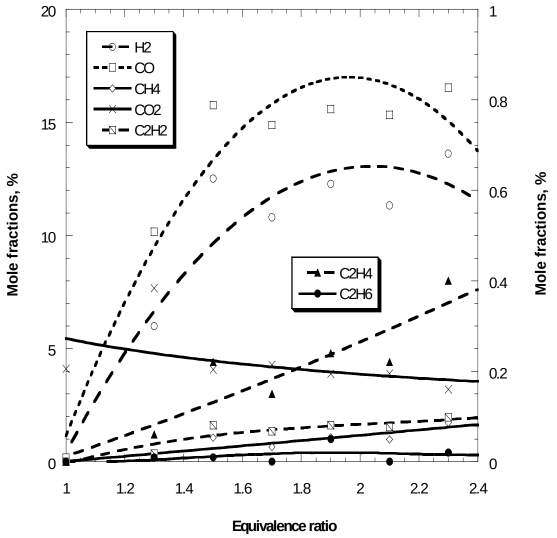

Fig. 7. Measured composition of chemical products as a function of equivalence ratio for rich and ultra-rich ethane-air mixtures.

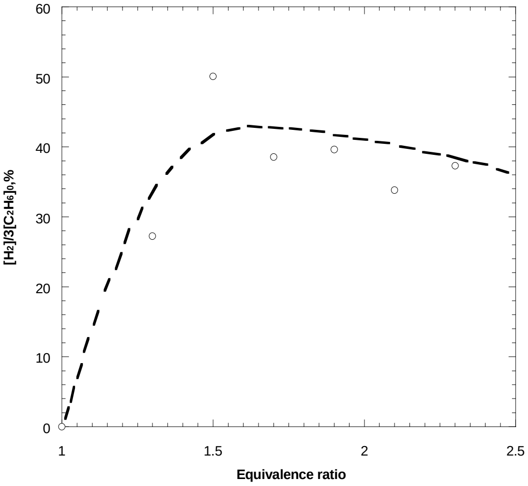

Fig. 8. Degree of ethane conversion to hydrogen for rich and ultra-rich wave.

3.2.3 Propane

Concentrations of CO_2 and H_2O decrease and partial oxidation products such as H_2, CO and significant concentrations of C_2 hydrocarbons appear in the exhaust gases (Fig. 9). H_2 and CO become dominant products for superadiabatic combustion waves at ϕ > 2.0. Unreacted C_3H_8 is measured in the products starting from ϕ = 1.5, generated at temperatures greater than 1500 K. However, during these tests the products temperature remained considerably above this critical temperature. Hence, the significant amounts of CH_4 and C_2H_2 species measured are due to finite rate kinetic effects, rather than the reverse steam reforming reaction. The measured concentrations of C_2H_4 and C_2H_6 could be as high as 0.2%. In fact, the ultra-rich superadiabatic wave is capable of inducing chemical transformation of propane in the mixtures with very small oxygen concentration. Thus, the process could be characterized as fuel reformation or modification rather than combustion. For example, Fig. 10 shows that the degree of propane conversion to hydrogen could be as high as 36% at the renge of equivalence ratios studied.

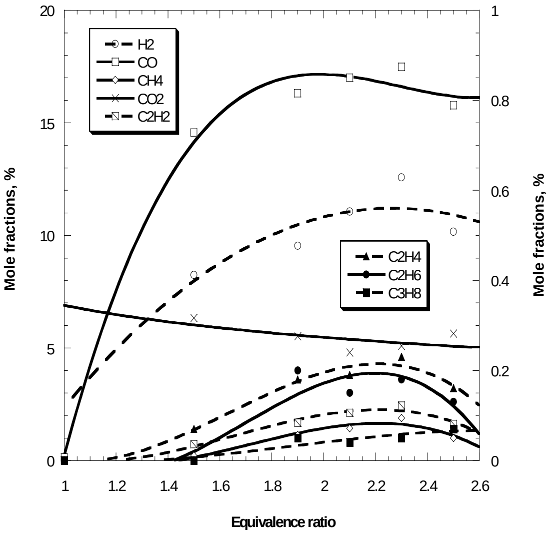

Fig. 9. Measured composition of chemical products as a function of equivalence ratio for rich and ultra-rich propane-air mixtures.

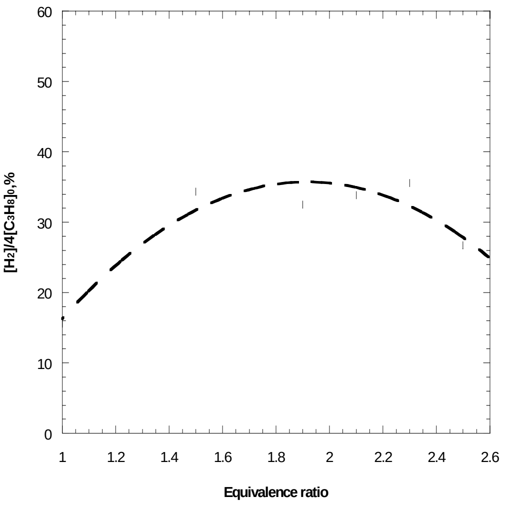

Fig. 10. Degree of propane conversion to hydrogen for rich and ultra-rich wave, u_g = 30 cm/s.

4. Conclusions

Filtration combustion waves formed in a packed bed of randomly arranged alumina pellets are studied experimentally for methane/air, ethane/air and propane/air mixtures in the range of equivalence ratios from 0.2 to 2.5. The studied range covers the superadiabatic and underadiabatic combustion waves formed in the lean and rich regions.

Downstream (superadiabatic) wave propagation was observed for ultralean (φ < 0.45) and ultrarich (ϕ > 1.5) methane-air mixtures. Upstream (underadiabatic) propagation corresponds to the range of equivalence ratios from 0.45 to 1.5. It was found that with equal heat content, rich mixtures have essentially higher combustion temperatures than corresponding lean mixtures. LVR of filtration gas combustion is observed for (CH_4)/air mixtures for all the range of ϕ.

For ethane-air mixtures, downstream (superadiabatic) wave propagation was observed for ultralean (ϕ < 0.4) and ultrarich (ϕ > 2.0) mixtures. Upstream (underadiabatic) propagation corresponds to the range of equivalence ratios from 0.4 to 1.9. Stable superadiabatic combustion of ultrarich mixtures was observed experimentally for the region of equivalence ratios above 2.0. The combustion temperature decreases in the range 0.9 < ϕ < 1.15 due to the transition to HVR of the wave propagation. The maximum combustion temperature corresponds to the ultrarich mixtures (ϕ ≥ 1.7); it is practically independent of the ethane content at this range.

For propane-air mixtures, downstream (superadiabatic) wave propagation was observed for ultralean (ϕ < 0.45) and ultrarich (ϕ > 2.0) mixtures. Upstream (underadiabatic) propagation corresponds to the range of equivalence ratios from 0.45 to 1.9. A maximum absolute value of velocity (0.01 m/s) is observed at the limit of LVR at $\phi = 0.9$. The maximum combustion temperature corresponding to the ultrarich mixtures ($\phi \geq 1.6$) where it is practically independent of the propane content.

In rich and ultra-rich mixtures complete combustion could not be achieved due to the low oxygen content in the mixture. This resulted in formation of partial oxidation products such as H_2, CO, and C_2 hydrocarbons. The maximum gernerated mole fraction of hydrogen was 14% for ethane, 13% for propane and 11% for methane flames stabilized inside the porous burner. This corresponds to a conversion to CO and H_2 of 70%, 55%, and 60% respectively. Considerable amounts of CH_4, C_2H_2 and C_2H_4 were measured in the combustion products of methane, ethane and propane at higher equivalence ratios due to finite chemical reaction rates. These results show that steady rich combustion inside a porous medium can be used to reform gaseous fuels into hydrogen and syngas.

A variety of fuels can be presently used in practical combustion applications both for the ultra-lean and ultra-rich cases. For example, natural gas compositions typically contain various concentrations of C1-C3 hydrocarbons. This suggests that further experimental and numerical studies of porous combustion of C1-C3 hydrocarbons and their mixtures are practically important. The further extension of this work will be experimental studies on porous combustion of characteristic hydrocarbon mixtures typical for various sources such as gasification, biogasification, natural gas suppliers and refineries. The numerical model describing the porous combustion of C1-C3 hydrocarbons and their mixtures will be developed and validated in comparison with experimental data.

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