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Full Length Article

The comparative and combined effects of hydrogen addition on the laminar burning velocities of methane and its blends with ethane and propane

Elna J.K. Nilsson [a,*], Astrid van Sprang [b], Jenny Larfeldt [c], Alexander A. Konnov [a]

[a] Combustion Physics, Lund University, P.O. Box 118, SE-221 00 Lund, Sweden
[b] Combustion Technology Section, Mechanical Engineering, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands
[c] Siemens Industrial Turbomachinery AB, SE-61283 Finspång, Sweden

HIGHLIGHTS

- Enhancement of laminar burning velocities of methane/ethane/propane as a result of H_2 addition.
- Hydrogen promotes SL of methane more than of blends of methane with ethane and/or propane.
- Increased reactivity due to hydrogen is a result of increased radical concentrations, in particular H.

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ABSTRACT

Laminar burning velocities of hydrocarbon blends of relevance to natural gas combustion, with addition of 0, 10, 35 and 50% hydrogen, were measured using the heat flux method. Hydrocarbon blends were methane (80%)/ethane (20%), methane (80%)/propane (20%) and methane (80%)/ethane (10%)/propane (10%), and in addition experiments were performed using pure methane as a fuel. For the first time it was shown experimentally that hydrogen promotes laminar burning velocity of blends with heavier hydrocarbons to a smaller extent than the well-studied effect on methane. Measurements show that enrichment by hydrogen results in 20–40% lower increase in laminar burning velocity for hydrocarbon blends compared to pure methane, depending on stoichiometry. Modeling points at the importance of increasing concentrations of OH, O and H radicals in H_2 enriched flames. At lean conditions increase in H atom concentration is of particular importance. The results are rationalized based on asymptotic flame theory analysis.

1. Introduction

Changes in fuel supply and natural gas variability strongly motivate development of fuel flexible gas turbines. The economic interests are in high combustion efficiency and security of fuel supply, and additional constraints are regulations enforcing low pollutants formation. Even though economic and environmental concerns push the development and implementation of renewable fuels, natural gas is expected to be the dominating fuel in gas turbine combustion for decades ahead, but likely to an increasing extent in blends with various other fuels. Energy producers demand systems that can run on various industrial by-product gases, ranging from heavy hydrocarbons to small and reactive hydrogen, H_2, and syngas, CO/H_2, fuels. Natural gases themselves mainly consist of alkanes, of which methane, CH_4, is the dominating component, commonly in the range of 80–95% of the total hydrocarbon content depending on the origin. Heavier hydrocarbons are mostly ethane, C_2H_6, and propane, C_3H_8, while C_4 and higher components are present in significantly smaller amounts.

Blending natural gas with hydrogen is of interest both because it is considered an environmental friendly fuel and due to the fact that it is often present in various process waste gases. Hydrogen is reactive with a high flame speed leading to increased risk of flash back when introducing it into burners originally designed for natural gas. The high reactivity has on the other hand a positive side in that H_2 has stabilizing effect on lean natural gas flames, used in, for instance, gas turbine combustion systems. Addition of H_2 has been shown to extend flammability range of methane/air flames in gas turbine burners [1] as well as in laboratory burners, as exemplified

* Corresponding author.
E-mail address: elna.heimdal_nilsson@forbrf.lth.se (E.J.K. Nilsson).

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in [2,3]. A fundamental parameter for assessment of flame stability, and therefore of interest for combustor operability, is laminar burning velocity, S_L, the speed at which a flame propagates through a combustible mixture. This parameter is also extensively used for validation of chemical kinetics mechanisms used in numerical models of combustion fluid dynamics which supports combustor designs.

Combustion of the main components of natural gas: methane, ethane and propane has been investigated in numerous publications; a complete overview, however, is beyond the scope of the present study. Laminar burning velocities of methane/air flames have been measured in a wide range of temperatures and pressures, as reviewed by, among others, Konnov [4] and Ranzi et al. [5]. As examples, recent and reliable data have been obtained from spherically expanding flames [6], counterflow flames [7] and burner stabilized flames [8]. Ethane and propane have been studied as pure compounds in flames with air [9,10]. It is well established, e.g., [9] that ethane is the compound with highest laminar burning velocity, burning 4–8 cm/s faster than methane/air at standard conditions, while propane is in between methane and ethane with the laminar burning velocities close to those of ethane at lean conditions, but closer to those of methane at rich conditions.

Fuel blends of methane with heavier hydrocarbons have also been extensively investigated in literature to elucidate the effect of natural gas composition on the burning velocity. Laminar burning velocities of the mixtures of methane with 20% of ethane have been determined using the heat flux method at atmospheric pressure [9] and spherical flame method at atmospheric and elevated pressures [6,11]. Lowry et al. [6] also measured laminar burning velocities of methane (80%)/propane (20%) fuel mixtures. It was shown in these studies that addition of the heavier hydrocarbons within the range expected for natural gas (<20%) yields higher laminar burning velocity by a few cm/s in comparison to flames of methane/air at standard conditions. From experimental and computational study on methane/ethane fuel blends Ravi et al. [11] concluded that increase in the laminar burning velocity compared to that for methane was mainly a kinetic effect.

The effect of H_2 enrichment has mainly been investigated for methane/H_2/air flames, as outlined in our previous studies [12,13] and summarized by Tang et al. [14]. Methane/air flames with addition of up to 68% vol. hydrogen were studied by Dirrenberger et al. [9] using the heat flux method. Donohue et al. [15] used the spherically propagating flame method to investigate mixtures of methane and H_2 at higher hydrogen fractions. From these studies it was concluded that for 50% H_2 with methane the laminar burning velocity was closer to that of pure methane than that of pure H_2.

Early measurements of laminar burning velocities of methane/air and propane/air flames with H_2 addition were reported for stoichiometric conditions by Milton and Keck [16], and over a wide range of mixture compositions by Yu et al. [17]. Tang et al. [14] pointed out that heavier hydrocarbons with addition of H_2 behave differently from blends with methane, in the sense that the combustion was enhanced to a smaller extent. The general knowledge on hydrogen addition to flames of one component fuels of heavier hydrocarbons was reviewed by Tang et al. [14].

Laminar burning velocities for blends of methane with heavier hydrocarbons and H_2 have not been measured. It is thus not clear to which extent H_2 promotes reactivity of hydrocarbon blends, in comparison to the effect on the pure fuel compounds. Several authors have quantified laminar burning velocities of natural gas enriched with hydrogen [15,18–22]. These studies do, however, use natural gas blends of various compositions and none of them attempt to investigate possible influence of variations in fuel hydrocarbon composition.

Understanding of the mechanisms underlying the laminar burning velocity enhancement related to hydrogen addition has been gained in a combined experimental and modeling study on butane/air flames, by Tang et al. [14]. The study also includes modeling of methane, propane and ethylene flames with H_2 addition. It was found that the kinetic effect is highly important, with minor contribution from thermal effect and negligible effect of diffusion. Modeling indicated that the promoting effect of H_2 should be strongest on methane, the least reactive of the studied fuels. In a modeling study by Brower et al. [23] effect of hydrogen addition on laminar burning velocities of a natural gas fuel with close to 20% heavier hydrocarbons was compared to pure methane fuel. The results indicate a more significant promoting effect of hydrogen on methane/air flame compared to methane/hydrocarbons/air flame. They also found that at large H_2 fractions reactivity of rich mixtures increased more than that of lean mixtures.

Further understanding of the chemistry of methane/hydrocarbon/H_2 flames can be deduced from different types of experiments reported in literature. Laboratory studies of laminar flames [24–26] and oxidation in Jet Stirred Reactor (JSR) [27,28] have been performed to investigate the promoting effect of hydrogen on hydrocarbon combustion. A study of a flat, premixed laminar flame [24] showed that enrichment by H_2 promotes reactivity leading to that peak heat release rate occurs at lower temperatures than for corresponding methane/air flames. Also, the thickness of the reaction zone was shown to decrease as H_2 fraction increased. Experimental flame structure at low [26] and atmospheric [25] pressures in combination with chemical kinetics modeling enabled an insight into the main reaction paths in natural gas flames with and without hydrogen admixture. An important conclusion was that the radical of largest importance for hydrogen abstraction reaction changed upon H_2 addition; in natural gas/air flames was OH, while H atoms became of significantly higher importance in the H_2 enriched flames.

Dagaut and co-workers performed oxidation experiments in a JSR on natural gas [27] and methane/ethane (10:1) mixtures [28] at lean and stoichiometric conditions, with various amounts of H_2. They demonstrated a significantly increased reactivity as a result of hydrogen addition, to a larger extent for lean mixtures than for stoichiometric. Using chemical kinetics modeling they inferred that the promoting effect to a large extent was a result of increased production of OH.

As evident from several experimental and simulation studies, addition of hydrogen to hydrocarbon/air flames results in an increased laminar burning velocity. The effect seems to be larger for methane compared to the heavier alkanes, or hydrocarbon mixtures like natural gas, but this far these trends have not been proved experimentally or quantified for individual methane/hydrocarbon fuel blends. The aim of the present study was to investigate the role of hydrocarbon composition of natural gas like blends, on sensitivity to H_2 enrichment. Laminar burning velocities are determined for mixtures of methane with ethane and propane, with addition of 0, 10, 35 and 50% of H_2. Gas mixtures selected for the study are methane (80%)/ethane (20%), methane (80%)/propane (20%) and methane (80%)/ethane (10%)/propane (10%). To allow comparison and validation of the experimental procedure flames of pure methane as a fuel were also studied.

2. Experimental

The heat flux method is based on stabilization of a flat flame on a burner at adiabatic conditions, which allow direct determination of the adiabatic laminar burning velocity. Practically this is achieved by employing a temperature controlled burner plate that ensures that heat loss from the flame to the burner is compensated