Experimental and simulation studies on the influence of hydrogen addition on the lean flammability limits of methane/air mixtures

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1 Introduction

Fuel-lean combustion is an attractive technology, offering prospective low emissions and high efficiency. However, it is also prone to issues like combustion instability and flame extinction [1]. In this context, flammability limits are investigated to identify conditions where flame extinction occurs; different experimental devices can be employed for this [2,3,4,5,6]. It is found that different fuels feature different flammability limits. Two rather extreme fuels in this respect are methane and hydrogen. Hydrogen can sustain combustion in a much wider range of equivalence ratios than methane. On the other hand, methane is widely-used as a fuel, while hydrogen is not often used in practical applications, mainly due to its high flame speed. No emission of carbon monoxide (CO) or carbon dioxide (CO2) is formed during the combustion [1,7,8]. The challenge is the safety by the storage and transport. Therefore, the flammability of methane should be strongly improved with the addition of hydrogen. In previous research, the flame speed of methane/air mixture increased dramatically with addition of hydrogen [9,10]. Meanwhile, with the addition of hydrogen, the peak of the concentration of OH-radical was increased, the lean flammability limit was lower, and the emission of NOx was also lower [11,12].

The goal of this work is to assess the influence of hydrogen addition on the flammability limit of lean methane/air mixtures under both laminar and turbulent conditions.

2 Methodology

2.1 Experiments

Flammability limits are determined experimentally in a constant-volume combustion bomb (Fig. 1 and fig. 2) that is equipped with 4 radial fans for generation of isotropic turbulence. The turbulence level is proportional to the fan speed, which can be controlled between 0 and 5000 rpm [14]. The ignition source is a glow-plug (BorgWarner BERU System GmbH) in the center of the combustion bomb. In an experiment, the desired fuel/air premixture is filled into the chamber at atmospheric pressure, and the glow-plug is activated to ignite the mixture and initiate combustion. The resulting flame propagation or extinction in the chamber is monitored by recording the pressure trace via a pressure transducer (Kistler 601Bh). After an experiment, the chamber is repeatedly evacuated and flushed with air to expel any remaining exhaust gas; then, the next experiment is started. The chamber wall temperature is continuously monitored to prevent a gradual heat-up of the chamber between experiments. In our study, premixtures of methane/air and methane/hydrogen/air with CH_4/H_2=9/1 (mol/mol) are used as fuel. More precisely, the molar composition ($CH_4/H_2/O_2/N_2$) in stoichiometric methane/air mixture was 9.5/0/19/71.5 mol%, and in stoichiometric methane/hydrogen/air mixture it was 9.17/1.02/18.9/70.9 mol%.

A series of experiments was conducted, with variations of the fuel/air equivalence ratio and turbulence level. The maximum pressure is extracted in each experiment to decide between flame propagation and flame extinction.

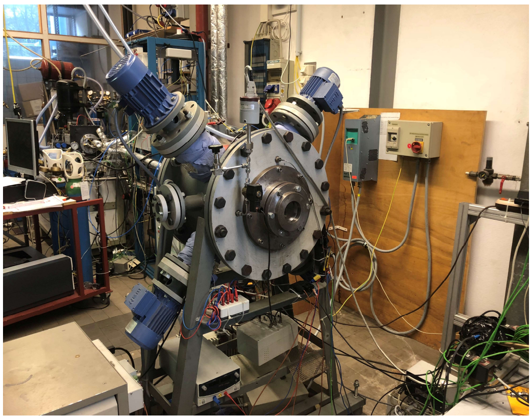

Fig.1: Experimental setup

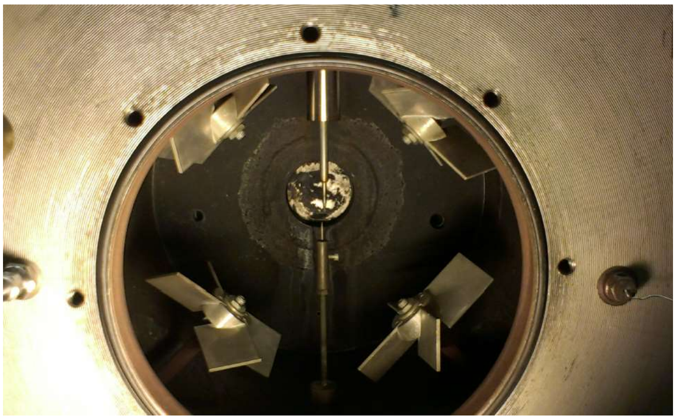

Fig.2: The fans and the glow-plug[15]

2.2 Simulations

Model simulations of flame initiation and propagation were performed using the in-house code INSFLA [13]. This code models flames in one-dimensional geometries, taking into account detailed chemistry and detailed molecular transport in the solution of the Navier Stokes equations. The simulation used a premixed counterflow configuration to compute the behavior of a small “sample” of fuel/air mixture. This sample is influenced by an externally imposed flow (as present by the turbulence in the experiment). We also assume that the sample just got into contact with the hot glow plug, by which it attains glow plug temperature (1700K) within some small region.

It is initialized with a one-dimensional temperature profile; on the left and right side of the domain ($r = 0$ m and $r = 0.1$ m), this gas has a temperature of 300K, corresponding to unburned, cold mixture. In the center, a temperature peak with 1700 K is imposed, representing the thermal initial condition imposed onto the gas by the hot glow-plug. Species composition and pressure profiles were set to the values of the experiment.

The counterflow configuration can be characterized by the tangential pressure gradient *J* over the domain [17]. In the counterflow, the fresh unburned gas flows towards the glow-plug because of the flow field (which is imposed in the simulation to mimick the flow induced by the fans in the actual experiment). Like in experiment, several simulations were performed to study the dependence of flame propagation on the composition (parameterized by the level of hydrogen addition and equivalence ratio), and the strength of the imposed flow field; the latter was used to mimick the effect of the turbulence in the experiment. Simulations were run until a steady state of the spatial profiles resulted. Using zero-gradient boundary conditions for species and temperature, the steady state allowed only two qualitatively different solutions: One where temperature on the boundaries corresponded the burning case (flame propagation), and one where it was equal to ambient (extinction). This outcome was recorded as a function of initial conditions (equivalence ratio, hydrogen addition, tangential pressure gradient).

3 Results

The results of the experiments are recorded using a binary model: “1” for a flame propagation; “0” for a flame extinction. Fig.3 shows all collected data points for the experiments with methane/air. The data show an overlap of points with flame propagation and extinction; they therefore indicate a transitional region instead of a “sharp” flammability limit. A Maximum Likelihood method was used to calculate the probability of flame propagation as a function of equivalence ratio; Fig. 4 shows an example of the resulting probability for methane/air at v’ = 0.35 m/s. The “overlap”, leading to a smooth transitional region rather than a sharp limit, is clearly visible.

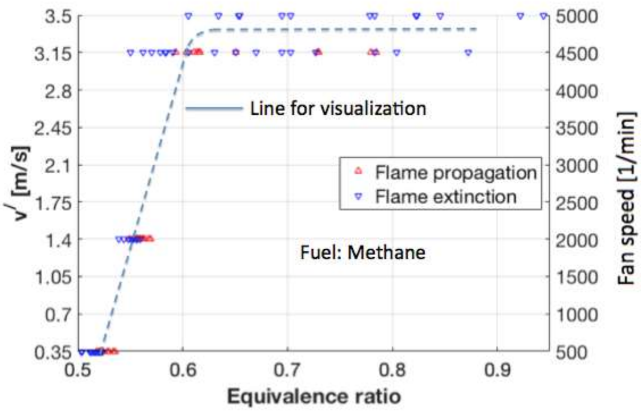

Fig.3: Data points for the experiments for methane/air mixture

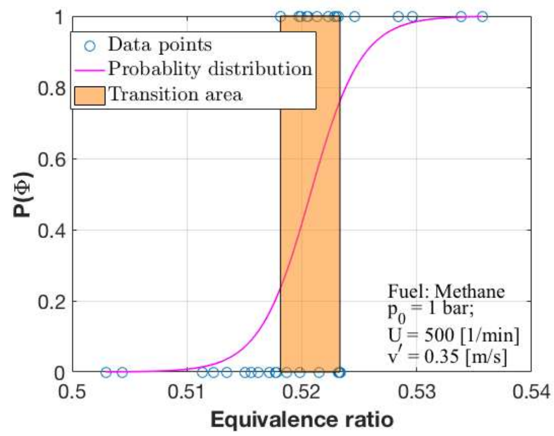

Fig.4: Probability distribution for methane/air mixture with weak turbulence (v’ = 0.35)

For pure methane, the flammability limit with no or only weak turbulence (v’ near 0.35 m/s) is near Φ = 0.52. If the turbulence is increased, the limit shifts towards stoichiometric. With turbulence v’ near 1.4 m/s, the limit is near Φ = 0.55. The width of the transitional region hardly changes, though. If the turbulence is increased further (v’ near 3 m/s) , the limit shifts strongly to Φ = 0.7, and the width of the transitional region now also widens strongly ($\Delta\Phi$ about 0.4). At this turbulence level, conditions are approached where the mixture cannot sustain flame propagation at all.

Addition of hydrogen shifts the flammability limit to smaller equivalence ratio. Like for methane/air mixture, the flammability limit of methane/hydrogen/air shifts towards stoichiometric with increasing turbulence. The transitional region with hydrogen addition is sharper throughout the whole investigated range of turbulence levels. For the highest investigated turbulence levels (v' = 3.5 m/s), the transitional region widens in comparison to smaller turbulence levels; it is however, much sharper than for methane/air mixture at v' = 3.15 m/s. No flame propagation is observed for the experiments for methane/air mixture at the turbulence level v' = 3.5 m/s; methane/hydrogen/air sustains combustion at this value of v'.

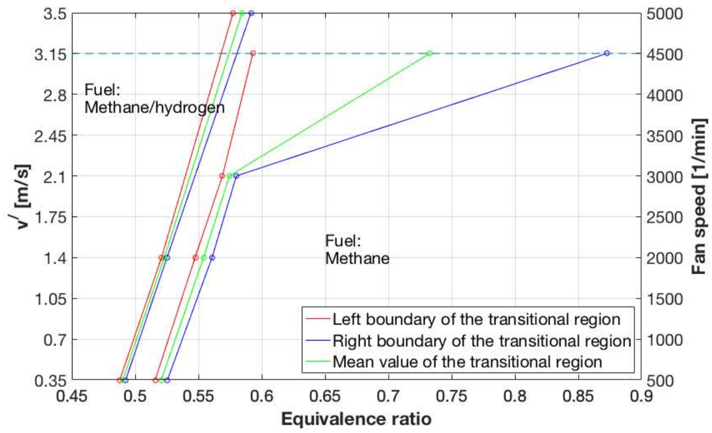

Fig.5: Transitional region for methane/hydrogen/air mixture and methane/air mixture

Simulations and experiments can be compared, at least on a semi-quantitative basis, if we employ the velocity gradient across the computational domain as a model parameter that captures the effect of turbulence intensity [7]. This is linked to the square root of the magnitude of J [17].

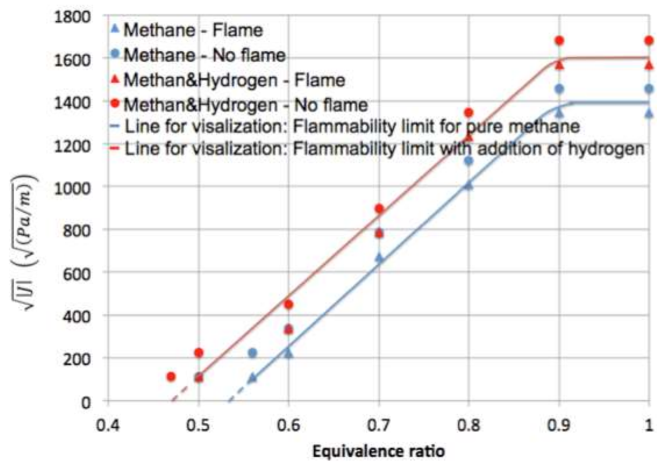

Fig.6: Simulation results of flammability limit for methane/air mixture and methane/hydrogen/air mixture

Fig.6 shows the simulation results in a diagram that, in this sense, is a simulation-based counterpart to the experimental data in Fig.5. Starting from very lean conditions, the flammability limit is shifting towards higher magnitudes of J as the equivalence ratio is increased. With hydrogen addition, the flammability limits widen significantly. At ϕ near 0.5, the methane/air mixture cannot be burned even at the weakest turbulence, while methane/hydrogen/air still burns with $|J| = 12600$ Pa/m.

While the absolute values of the extinction limits in ϕ are slightly shifted between experiment and simulations, experiment and model agree in that 10% hydrogen addition shifts the flammability limit to the left (towards smaller ϕ) by about $\Delta\phi$=0.05. The simulation model predicts the existence of a region where the flammability limit is nearly independent of ϕ, i.e., runs nearly horizontally in the diagrams. This is discernible also in our experiments for the case of methane/air. Simulations predict this region also for methane/hydrogen/air, but at larger magnitudes of J. To assess whether (and where) this region is also found in experiment would require measurements at turbulence levels above the ones admissible by our setup (due to limits of maximum allowable fan speed).

Literature reports that H2 has larger flame speed and wider flammability limits than CH4; our results are consistent in that the mixture CH4/H23 has wider limits than CH4. Part of this effect might also be attributed to the high diffusivity of hydrogen.

4 Conclusion

Experiments and simulations were used to study the influence of hydrogen addition onto the flammability limits of methane/air and methane/hydrogen/air mixtures under laminar and turbulent conditions. Addition of 10% hydrogen to methane/air generally shifts the lean flammability limit to smaller equivalence ratios. The transitional region between flame propagation and extinction was also sharper when hydrogen was added. At v' near 3.5, methane/air could not be ignited at all in our study, while methane/hydrogen/air showed a transitional region similar to lower turbulence.

In the simulation, the highest turbulence, with what the mixture can still be ignited, at different equivalence ratio is investigated. With the addition of hydrogen, the mixture can be ignited at higher turbulence, and the effect of hydrogen is greater, when the equivalence ratio is near stoichiometric. At equivalence ratio near 0.5, the methane/air mixture cannot be ignited even with the weakest turbulence, while the methane/hydrogen/air mixture can be ignited with $J = -12600\ Pa/m$. Experiments and simulations show that hydrogen addition widens the flammable range under both laminar and turbulent conditions. Meanwhile, the mixture with same equivalence ratio can be ignited at higher turbulence. The addition of hydrogen to lean methane/air mixture has a great effect for a larger flammability limit range and a more stable flame.

5 Acknowledgements

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References

[1] Dunn-Rankin, D. (2008). Lean Combustion. Lean Combustion.

[2] Coward, H. ., & Jones, G. . (1952). Limits of flammability of gases and vapors. Bulletin 503, U.S. Department of the Interior, Bureau of Mines.

[3] Shoshin, Y., Tecce, L., & Jarosinski, J. (2008). Experimental and computational study of lean limit methane-air flame propagating upward in a 24mm diameter tube. In Combustion Science and Technology (Vol. 180, pp. 1812–1828).

[4] Yamaoka, I., & Tsuji, H. (1979). An experimental study of flammability limits using counterflow flames. Symposium (International) on Combustion, 17(1), 843–855.

[5] Chen, Z. H., & Sohrab, S. H. (1995). Flammability limit and limit-temperature of counterflow lean methane-air flames. Combustion and Flame, 102(1-2), 193–199.

[6] Cashdollar, K. L., Zlochower, I. A., Green, G. M., Thomas, R. A., & Hertzberg, M. (2000). Flammability of methane, propane, and hydrogen gases. Journal of Loss Prevention in the Process Industries, 13(3-5), 327–340.

[7] Warnatz, J., Maas, U., & Dibble, R. W. (2006). Combustion: Physical and chemical fundamentals, modeling and simulation, experiments, pollutant formation. Combustion: Physical and Chemical Fundamentals, Modeling and Simulation, Experiments, Pollutant Formation.

[8] Joos, F. (2006). Technische Verbrennung: Verbrennungstechnik, Verbrennungsmodellierung, Emissionen (German Edition). Springer.

[9] Yu, G., Law, C. K., & Wu, C. K. (1986). Laminar flame speeds of hydrocarbon + air mixtures with hydrogen addition. Combustion and Flame, 63(3), 339–347.

[10] Ilbas, M., Crayford, A. P., Yilmaz, I., Bowen, P. J., & Syred, N. (2006). Laminar-burning velocities of hydrogen-air and hydrogen-methane-air mixtures: An experimental study. International Journal of Hydrogen Energy, 31(12), 1768–1779.

[11] Griebel, P., Boschek, E., & Jansohn, P. (2007). Lean Blowout Limits and NO_x Emissions of Turbulent, Lean Premixed, Hydrogen-Enriched Methane/Air Flames at High Pressure. Journal of Engineering for Gas Turbines and Power, 129(2), 404.

[12] Schefer, R. W. (2003). Hydrogen enrichment for improved lean flame stability. International Journal of Hydrogen Energy, 28(10), 1131–1141.

[13] U. Maas, J. Warnatz, Ignition processes in hydrogen-oxygen mixtures, Combustion and Flame. Volume 74, Issue 1, 1988, Pages 53-69, ISSN 0010-2180

[14] S. Lindenmaier, Zeitaufgelöste Laser-diagnostische Untersuchung der Funkenzündung. PhD thesis, 2001, Universität Stuttgart.

[15] Ulrich, J. Aufbau und Charakteristisierung eines Glükerzen-basierten Zündsystems für ein Experiment zur Untersuchung der Flammenlöschung. Bachelor thesis, 2016, Karlsruhe Institute of Technology

[16] Henning, R. Experimentelle Untersuchung und statistische Auswertung der mageren Flammenlöschgrenzen von Methan- und Ethano-Luftgemischen in isotroper Turbulenz. Bachelor thesis, 2016, Karlsruhe Institute of Technology.

[17] Stahl, G., & Warnatz, J. (1991). Numerical investigation of time-dependent properties and extinction of strained methane and propane-air flamelets. Combustion and Flame, 85(3-4), 285–299.