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# Combustion regimes of low stretched premixed methane-hydrogen-air mixtures near flammability limits

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Abstract. The experimental study of counterflow premixed low-stretched flames stabilized in a flat channel is described in the paper. A premixed methane-air mixture was investigated. The effect of the channel gap on the flammability limits was evaluated. It was found that increasing the channel gap expands the flammable region to a lean mixture area. The effect of the hydrogen addition on the flammability limits was also studied as part of the paper. This leads to lower Lewis numbers and various unstable combustion modes such as running waves, moving cells and sporadic flames. Similar modes have been observed previously in microgravity experiments. The obtained combustion modes were classified, and the area of premixed methane-hydrogen-air flames existence was determined. Additionally, the regime diagram for the mixture with hydrogen content of 50% by volume in the fuel was plotted as a results of this study.

**Keywords:** experiment, low stretched flame, hythane, methane-hydrogen-air mixtures, flammability limit, wrinkled flames, sporadic flames, cup-like flames.

#### 1. Introduction

Non-stationary and coherent combustion modes of gas-phase flames usually form near the flammability limit. Evolution of various instabilities related to thermal-diffusive instability (TDI) could lead to non-plane and dynamic flame structures, such as cellular or sporadic flames [1]. Thermal-diffusive instability dominants at ultra-lean mixtures with low Lewis number [2]. Hydrogen addition to methane-air mixture allows reducing Lewis number [3]. Such fuel mixture was named Hythane, and its study is highly promising as a bridge to eco-friendly hydrogen energy. Besides that, TDI-induced combustion modes often appear outside of continuous flame front extinction limit, and their study is necessary for fire and explosion safety [1].

Experimental study of near-limit combustion regimes requires expensive experimental facilities to create microgravity conditions (such as "drop towers" [4], parabolic flights [1, 2, 5], and space experiments [6]) due to the TDI-induced combustion modes appearance at ultra-low mixture flows. Currently there is a deficit of experimental data related to dynamic behavior and structure of premixed near-limit flames obtained under laboratory conditions.

Experimental setup consisting of two countercurrent slot-jet burners and two parallel quartz plates forming the flat channel was suggested in our previous studies [7, 8]. Designed setup allowed us to study low stretched flames in laboratory conditions [9]. Goal of this work was to study the near-limit low stretched premixed methane-hydrogen-air flames in the experiment.

#### 2. Experiment

Counterflow technique is widely used for experimental study of flammability limit and flame structure [1, 2, 4, 5]. Flow field of counterflow flames could be described by the stretch rate (velocity gradient) which is defined as:

$$a = V_0 / 0.5 \times L , \qquad (1)$$

where  $V_0$  is unburned mixture inlet velocity, L is the distance between burner nozzles.

Fuel content in mixture is defined by equivalence ratio:

$$\varphi = f/f_S , \qquad (2)$$

where f is actual fuel/oxidizer ratio,  $f_S$  is stoichiometric ratio between fuel and oxidizer.

# 2.1. Experimental setup

In this study we modified the experimental setup used in our previous work [7]. Two parallel quartz plates (plate size  $120 \times 50 \times 1$  mm) formed a flat channel and four steel bars mounted inside the channel formed the side walls of slot-jet burner nozzles as shown in Fig. 1. Near the nozzle's ends we mounted two porous copper bars to uniform the fresh mixture flow. 200 µm K-type thermocouple was inserted into the space between porous copper bars through the tiny hole in nozzle's side wall to measure the temperature of unburned mixture. Another K-type thermocouple was mounted outside of the bottom quartz plate at the center to determine the thermal stabilization of counterflow burner. Thermocouples were connected to the controller Thermodat 29M6 (Russia) to register temperature measurements. To create identical flows, burner's bodies were 3D-printed from ABS-plastic, and combustible mixture was supplied through the 4 mm inner diameter pipes of equal length. Gas flow rate was controlled by pre-calibrated mass flow controllers Bronkhorst F-series (Netherland). All connections were accurately sealed and checked for leakage. Thus, all modifications allowed us to remove the coolant system used in [7], minimize the heat loss to the burners, and maintain the same size of experimental area as in previous experiments [7]. Flame images and dynamic behavior were captured by two digital photo cameras Nikon D7200 (Japan) which were mounted at two points: above the burner to make photos or videos from the top view, and opposite to the burner to visualize the flame inside the gap between the quartz plates.

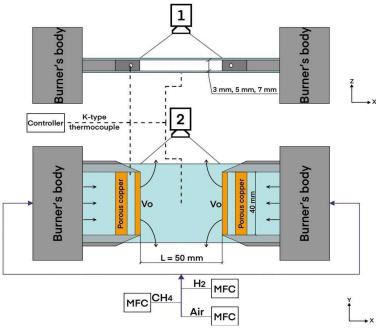


Fig. 1. The scheme of the experimental setup.

#### 2.2. Experimental technique

Lean methane/air and hythane/air mixtures were used during experiments. All combustible gases used in experiments had the purity more than 99.9%. For methane/air mixture combustion modes and flammability limit were obtained according to the procedure described below. Gas flow rates in accordance with predetermined stretch rate and equivalence ratio were set on mass flow controllers, and mixture was ignited from external heat source. After flame ignition, the experimental setup became thermally stabilized in several minutes. Stabilization was identified by constant value of measured temperatures of unburned mixture and quartz plate. After that, photos of observed stationary combustion mode or video recordings (in case of non-stationary regime) were captured. Inlet mixture temperature, atmospheric pressure, and gas flow rates registered in experimental record

to reproduce the actual value of stretch rate. Further, depending on situation, equivalence ratio, or stretch rate value, or both were decreased, another thermal stabilization regime was achieved, and procedure of data collection repeated. Approaching the flammability limit, we narrowed the decrement change of equivalence ratio and stretch rate.

For hythane/air mixture we used the same method, but equivalence ratio and stretch rate value were changed according to previously established protocol. This was necessary to ensure that the hydrogen concentration in the fuel remained constant. Hydrogen concentration in fuel was defined as:

$$x = \frac{V_{H2}}{V_{H2} + V_{CH4}},$$
(3)

where  $V_{H2}$  is hydrogen volume rate and  $V_{CH4}$  is methane volume rate.

Analysis of experimental data included determination of the actual flow rates of the fresh mixture based on the temperature and atmospheric pressure, and averaging of the extinction points based on the results of three measurements. Based on the results of data analysis, a regime diagram was compiled in the plane stretch rate/equivalence ratio.

# 3. Results and discussion

#### 3.1. Effect of channel gap on flammability limit of methane-air flames

Lean  $0.55 \le \varphi \le 0.8$  methane-air counterflow flames were studied in the channels with 3 mm, 5 mm, and 7 mm gap for stretch rate range from 5 s<sup>-1</sup> to 45 s<sup>-1</sup>. Flammability limits obtained for different channel's gap presented on stretch rate/equivalence ratio diagram shown in Fig. 2. Blue markers indicate flammability limit for 7 mm channel, red markers – for 5 mm channel, and green markers – for 3 mm channel respectively. Reducing the channel's gap leads to the narrowing of flammable region in all range of stretch rates and equivalence ratios (Fig. 2). Near stagnation flames (NSF) and distant flame (DF) [7] regimes were observed for 5 mm and 7 mm channels. NSF area is located inside of upper peninsula in the range of stretch rate from 20 s<sup>-1</sup> to 45 s<sup>-1</sup> for 5 mm channel and from 22 s<sup>-1</sup> to 45 s<sup>-1</sup> for 7 mm channel. DF area expanded to the lean region with reducing of stretch rate.

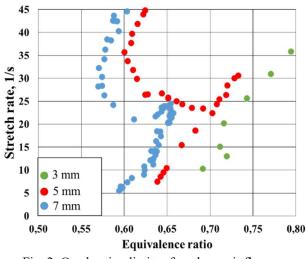


Fig. 2. Combustion limits of methane-air flames.

Notably, only DF combustion mode was found for 3 mm channel in studied stretch rate range. Obtained results are in qualitative agreement with theoretical prediction described in our previous paper [8]. Moreover, experiments showed that channel increase allowed us to achieve lower values

of stretch rate. For 3 mm channel, flame fronts stabilized on nozzle's surface at stretch rates lower than 10 s<sup>-1</sup>. Whereas, for 5 mm and 7 mm channel, critical stretch rate values were 7 s<sup>-1</sup> and 5 s<sup>-1</sup> respectively. Therefore, all further experiments we conducted with 7 mm channel.

#### 3.2. Effect of hydrogen addition on flammability limit

To investigate the impact of adding hydrogen to the fuel, we utilized two types of hythane/air mixtures: Hythane30, which contains 30% hydrogen by volume, and Hythane50, which contains 50% hydrogen. Fig. 3 demonstrates the effects of hydrogen addition on the flammability limits in 7 mm channel. Blue markers indicate flammability limit of methane/air mixture, yellow markers – Hythane30/air mixture, and burgundy markers – Hythane50/air mixture. Fig. 3 clearly shows how hydrogen addition significantly effects the flammability limit, shifting the extinction points to lean region. As it can be also seen, NSF region (upper peninsula of the plot) expands with the increase of hydrogen content in fuel. It means that with hydrogen addition  $\varepsilon$ -shape flammability limit curve [7] lead to C-shape curve [4] typical for conventional stretch flames. Hence, we can assume that increase of hydrogen content in the fuel leads to a decrease of the interface heat transfer intensity and minimization of channel's wall effect, respectively. Based on this, we studied the combustion limit for Hythane50/air flame in more detail and obtained flame quenching related to the heat loss to the burner's nozzle. It was observed that in equivalence ratio from 0.53 to 0.45 flame extinction points smoothly shift from stretch rate value 2 s<sup>-1</sup> to 4 s<sup>-1</sup> (see Fig. 3).

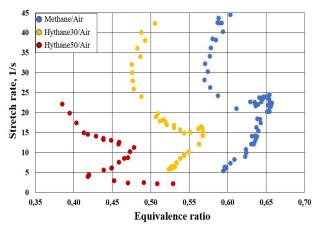


Fig. 3. Flammability limits of methane-hydrogen-air flames and methane-air flames in 7 mm channel.

In addition to the flammability limits, we also studied combustion regimes inside the combustion region. It was found that for Hythane30/air mixture only stable modes (NSF and DF) exist in the studied region, same as for methane/air mixture. It means that addition of 30% of hydrogen to the fuel is not enough to decrease the effective Lewis number at which the effects related to thermal-diffusive instability will appear. However, for Hythane50/air mixture such effects were found. Detailed description of near-limit combustion modes obtained for Hythane50/air mixture is given in the next section.

3.3. Combustion modes of methane-hydrogen-air flames with 50% hydrogen addition by volume Fig. 4 illustrates the typical combustion modes, observed in experiments with Hythane50/air mixture. NSF mode (Fig. 4a) existed in a wide range of stretch rate and equivalence ratio inside of upper peninsula of flammability limit curve. Since we were interested in unstable regimes, we focused on stretch rate region less than 10 s<sup>-1</sup>. At equivalence ratio  $\phi = 0.55$  burner-stabilized flat flame was obtained (Fig. 4b). This combustion mode is characterized by relatively high normal velocity to be the limiting case of DF mode. With the decrease of equivalence ratio multiple cells occurred on the flame surface (Fig. 4c). It was found that cell size and quantity strongly depend on mixture content. The higher equivalence ratio, the larger number of cells appears, and the size of each cell is smaller. With the decrease of equivalence ratio, cellular flames are stabilizing further from the burner surface, and the number of cells reduce to two. Subsequent mixture leaning shifted flame fronts closer to the stagnation zone and cellular mode transformed to a non-stationary sporadic regime (Fig. 4d) [1]. After sporadic regime, flame quenching was observed. At stretch rates less than 5 s<sup>-1</sup>, two more unstable modes were found: traveling wave (Fig. 4e) and moving cells (Fig. 4f). At wave mode wrinkled flame front stabilized near the burner surface, and small wave periodically propagated on the flame front from the center to the edges. Moving cells mode was the similar to the traveling wave, except that continuous flame front in this regime was separated to small cup-like cells, which moved from the center to the edges of burner surface.

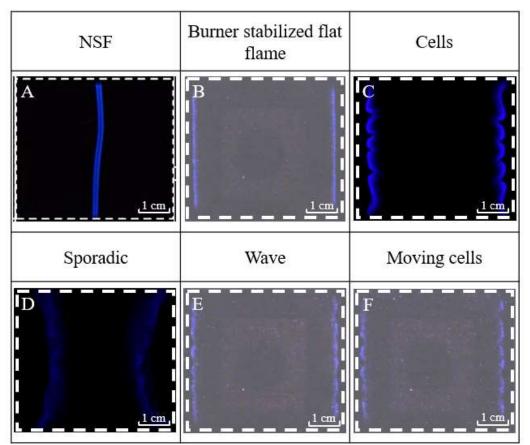
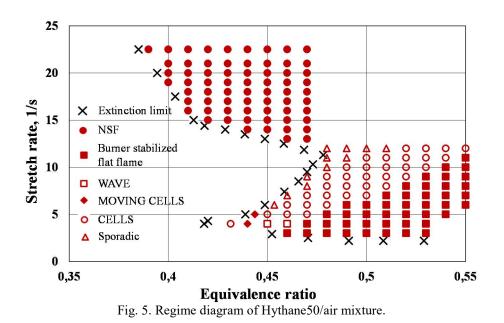


Fig. 4. Combustion modes observed for Hythane50/air mixture.

All obtained combustion modes were plotted at regime diagram presented in Fig. 5. Black crosses indicate the extinction points, solid circles – NSF, solid squares – burner-stabilized flat flame, rings – cellular flame, triangles – sporadic flame, hollow squares are traveling waves, and solid rhombs – moving cells, respectively. Fig. 5 demonstrates that burner-stabilized flat flame mode expands with the decrease of stretch rate and equivalence ratio. At the same time, sporadic and cellular flame region became narrower. Sporadic regime was not observed at the stretch rate value less than 6 s<sup>-1</sup>, but traveling wave and moving cells modes appeared. Notably, combustion regimes boundaries plotted in diagram are nominal, because combustion modes transformed smoothly. In conclusion, our study demonstrates all combustion modes previously observed only in the microgravity conditions during parabolic flights [1, 2]. Moreover, there is an opportunity to study in detail thermal-diffusive effects, occurring in gas combustion under Earth gravity conditions.



#### 4. Conclusion

The study investigated the flammability limits of low-stretched premixed methane-air flames in flat horizontal channels of different heights. Also, flammability limits and combustion modes of premixed methane-hydrogen-air flames with hydrogen concentrations of 30% and 50% by volume were analyzed in a 7 mm channel. The obtained combustion limit curve tends to be the C-shaped one with an increase of hydrogen concentration in the fuel. This also leads to a decrease of the Lewis number in the combustible mixture and allowed for the observation of the unstable combustion modes such as running waves, moving cells, and sporadic flames, which were previously observed in microgravity experiments. The results of the study demonstrate the possibility of observing TDI-induced combustion modes for an extended period without the need for microgravity conditions. Consequently, this opens up the potential for using a wide range of previously unavailable experimental methods with the proposed laboratory setup for a more detailed study of the structure and properties of the obtained combustion modes.

Based on the experimental data, the authors suggest that further research using the proposed experimental configuration is promising. In addition, further increase of the hydrogen content in the mixture and decrease of the stretch rate could allow obtaining combustion modes in the form of isolated sources of energy release with a dominant diffusive transport mechanism, such as flame balls.

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