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Review

# Physical and Chemical Features of Hydrogen Combustion and their Influence on the Characteristics of Gas Turbine Combustion Chambers

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Abstract: Hydrogen plays a key role in the transition to a carbon-free economy. One of the areas of hydrogen use is the substitution of hydrocarbon fuel in gas turbine engines and power plants. This review article discusses the features of hydrogen combustion and their influence on the characteristics of combustion chambers in comparison with methane. The paper presents both the results of a study of pure hydrogen or methane and methane-hydrogen mixtures with different hydrogen content. Among the main features, it is worth noting a smaller ignition delay time and higher laminar flame speed with a shift in its maximum value to a rich mixture, which significantly affects on flashback inside burners premixer, especially at elevated air temperatures. Another feature is the increased temperature of the flame, which can lead to an increase in the rate of nitrogen oxides formation. However, wider combustion concentration ranges contribute to the stable combustion of hydrogen at temperatures lower than those of methane. Along with this, it was shown that even at the same adiabatic temperature, more nitrogen oxides are formed in a hydrogen flame than in a methane flame, which indicates an additional mechanism for NOx formation in addition to the Zeldovich mechanism. The article also summarizes some of the results of a study on the effect of hydrogen on the occurrence of thermoacoustic instability, which depends on the initial nature of pulsations during methane combustion. The presented data will be useful both to engineers who are engaged in solving the problems of designing hydrogen combustion devices, and to scientists in this field of study.

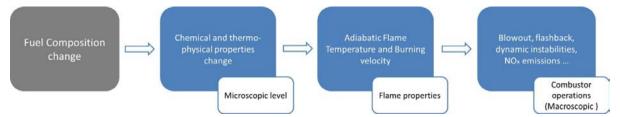
Keywords: hydrogen; combustion chamber; emission; gas turbine engines; flame blowout

# INTRODUCTION

The raising importance of using hydrogen (H<sub>2</sub>) as a fuel for heat engines and power plants is associated with the need to switch to carbon-free technologies, which in the long term is intended to solve several problems associated with global warming [1-2]. In addition, the use of hydrogen as a fuel makes it possible to increase the theoretical power limit of gas turbine engines and reduce the dependence of aircraft and industrial plants on fossil fuels.

In [3], a detailed analysis of the influence of the composition of gaseous fuel, in particular CO, methane, natural gas, syngas and hydrogen, on the parameters of the combustion process and the characteristics of gas turbine combustors was given. The logical structure of the analysis is shown in Figure 1. Changing the composition of the fuel leads to a change in its chemical and physical characteristics, which in turn affects the properties of the flame (flame temperature, ignition delay time, flame speed) and will determine the main characteristics of the combustion chamber (pollutant emission, combustion efficiency, static and dynamic stability, flashback). The purpose of this work is to analyze the

literature in the field of hydrogen combustion and its effect on the characteristics of combustion chambers in comparison with the widely used hydrocarbon fuel – natural gas.



**Figure 1.** - Scheme of fuel composition influence analysis [3].

# 1. Features of the processes of hydrogen combustion

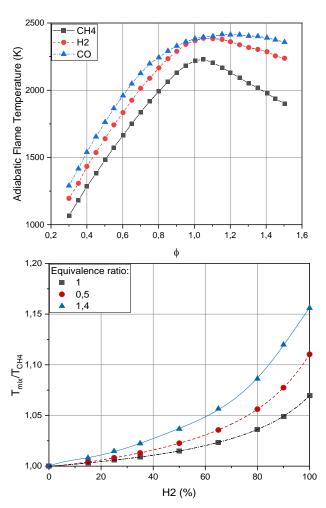
# 1.1. Properties of hydrogen and flame temperature

The use of hydrogen in gas turbines is mainly considered as a replacement for the widely used natural gas (due to the similarity of properties in this paper, natural gas and methane are considered as similar fuels). At the same time, at the first stage of technological re-equipment, the partial or complete replacement of natural gas with hydrogen without significant construction changes is considered. This possibility can be assessed at the stage of comparing the physicochemical properties of hydrogen with natural gas. Table 1 shows that hydrogen has a significantly lower density (about 8 times lower than methane), wider ignition concentration limits, and, in general, is a more reactive fuel. The net calorific value per unit mass is more than three times higher that for methane, but per unit volume it is almost 2.5 times lower. Consequently, at the same thermal power, the required volumetric flow rate of hydrogen is more than twice the flow rate of methane.

<b>Table 1.</b> - Properties of	hydrogen	compared to	o the natura	I gas [4	-7]
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	Hydrogen	Natural gas
Density at 273 K, kg/m <sup>3</sup>	0,09	0,72
Molar mass, kg/kmol	2,016	16,04
Ignition limits (Φ)	0,1-7,1	0,4-1,6
Low calorific value, MJ/kg	120	50
Low calorific value, MJ/m <sup>3</sup>	11	36
Maximum laminar flame speed, m/s	3,25	0,45
Adiabatic flame temperature at $\Phi = 1$ , K	2402	2216
Stoichiometric coefficient	34,2	17,2
Melting temperature, K	14	91
Boiling temperature, K	21	112
Critical temperature, K	33	191
Critical pressure, amt	12,8	45,2
Autoignition temperature, K	850	810

Hydrogen also has a higher adiabatic flame temperature. This can increase the probability of overheating of the elements of the combustion chamber and the turbine while maintaining the ratio of excess fuel in the combustion zone. The influence of the content of hydrogen in its mixture with natural gas on the flame temperature is shown in Figure 2. The flame temperature is mainly affected by the specific heat of combustion and flame speed. Since they are relatively high for hydrogen, during its combustion, while maintaining the stoichiometric coefficient, the flame temperature will be higher. It is worth noting that the relative increase in the temperature of the flame of hydrogen compared to methane is greater for rich mixtures (about +15-17%), slightly less for lean mixtures (+10-12%) and least of all for stoichiometric mixtures (about 7-8%). Thus, it can be said that the heat load on the flame tube head of the combustion chamber can become 10-15% higher when switching to hydrogen fuel, which will require additional affort to ensure thermal state.

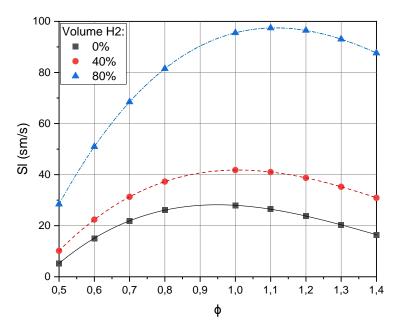


**Figure 2.** – Influence of hydrogen volume concentration on the normalized temperature of the methane-air mixture flame [3, 8, 9].

### 1.2. Laminar flame propagation speed

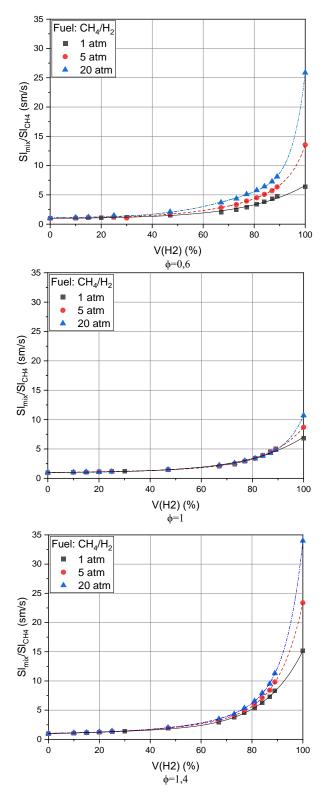
One of the main properties of a laminar flame is the propagation speed. Figure 3 shows the dependence of the laminar flame propagation speed on the equivalence ratio. The first thing worth noting is that the maximum propagation speed of a laminar hydrogen flame is several times higher for methane (100-120 m/s compared to 30-40 m/s under normal atmospheric conditions). The second feature is the shift of the maximum value to the region of rich mixtures. The maximum value of the flame speed of the hydrogen mixture shifts to the region  $\phi$ =1.2–1.3 (Figure 3), which approximately corresponds to the position of the flame temperature maximum. For combustion chambers, this may mean that

rich mixtures will burn more intensively, increasing the probability of flashback. However, hydrogen combustion can occur at higher flow rates without loss of combustion efficiency.



**Figure 3.** – Influence of the molar fraction of hydrogen on the flame progragation speed of methane-hydrogen mixture at atmospheric pressure and 300 K [9, 10].

The effect of pressure on the relative laminar flame speed of methane-hydrogen mixtures is shown in Figure 4.



**Figure 4.** - Dependence of the propagation laminar flame speed of methane-hydrogen mixture on the volume concentration of hydrogen [9, 11, 12].

The addition of hydrogen has the greatest effect on the flame speed of rich mixtures at high pressures (an increase in speed by a factor of ~35 compared to methane laminar flame speed at a pressure of 20 atm). For lean mixtures, the effect of hydrogen is somewhat less compared to rich mixtures, by about 25 times. In general, it can be noted that there is an effect of the influence of hydrogen additions on the flame speed, which largely depends on pressure and increases with its increase. This complicates the process of extrapolating

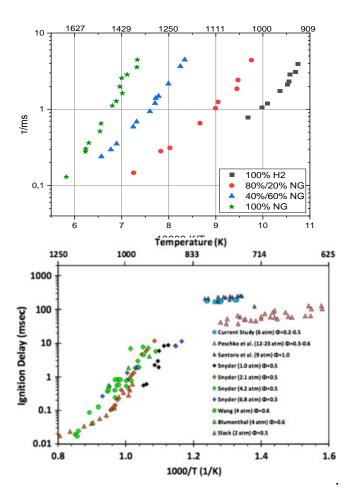
the results obtained from atmospheric studies to real conditions in gas turbine engines, which leads to the need for additional experimental development of the working process when hydrogen is burned in combustion chambers at elevated pressures, and the feasibility of such studies is much higher than when burning natural gas.

In this case, for any composition, the following regularity can be noted. The increase in normal flame propagation speed can be divided into three modes. At a hydrogen content of up to 50-60%, the increase in the laminar flame speed with an increase in the proportion of hydrogen is approximately linear and does not exceed 25%, and it is believed that hydrocarbons predominate in the chemical composition. At the highest levels of hydrogen content (>75-90%) there is an exponential increase in flame speed due to the dominant effect of hydrogen oxidation chemistry - the flame is considered predominantly hydrogen.

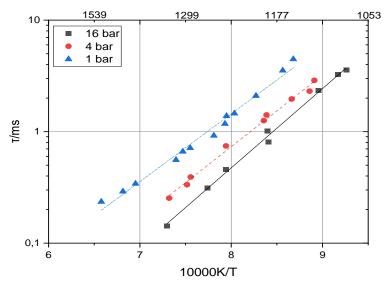
### 1.3. Ignition delay time and autoignition temperature

One of the most important characteristics of the combustion process is the ignition delay time. In combustion chambers, this value affects the amount of time that is available for pre-mixing the fuel with air before ignition and combustion (when the air temperature after the compressor is higher than the auto-ignition temperature of the mixture). On the one hand, to achieve a low level of nitrogen oxide emissions, a high quality of fuel-air mixture preparation is required, on the other hand, the premix burner section (premixer) must be designed in such a way as to avoid undesirable ignition in it. Due to the high temperature and pressure at the inlet to the combustion chambers of gas turbines, this becomes a difficult task, and for highly reactive fuels such as hydrogen, it is even more difficult.

Figure 5 shows that the ignition delay time strongly depends on the temperature and composition of fuel-air mixtures: the ignition delay time decreases by orders of magnitude with an increase in the proportion of hydrogen in the fuel. Similar results were obtained in [8]. Despite this, at typical air temperatures at the inlet to the combustion chamber (600-800 K), the ignition delay time for lean mixtures of hydrogen with air is about 100 ms, which is quite enough to organize the mixing process. It was shown in [14-16] that an increase in pressure also leads to a decrease in the ignition delay time of the methane-hydrogen mixture, as well as for pure methane (Fig. 6).



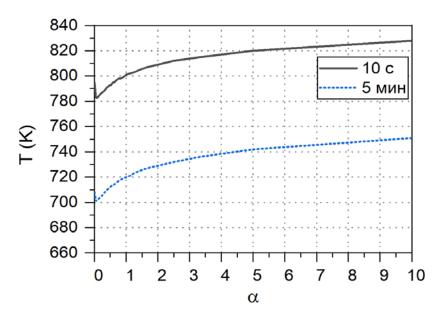
**Figure 5.** - Change in the ignition delay time of a mixture of natural gas depending on the content of hydrogen [14-16].



**Figure 6.** - Change in ignition delay time depending on pressure (experiment  $\Phi$  =0.5, mixture of 40% H2 and 60% natural gas) [15].

According to our estimate (using a perfectly mixed reactor), the minimum temperature of autoignition of hydrogen under atmospheric conditions with a residence time in the reactor of  $10 \, \mathrm{s}$  is about  $780-810 \, \mathrm{K}$  (figure 7), according to [10, 17] (5 minutes) it is about  $700 \, \mathrm{K}$ . For stoichiometric mixtures of methane - about  $850 \, \mathrm{K}$ . The minimum autoignition temperature shifts to the region of rich mixtures and is approximately  $70-150 \, \mathrm{degrees}$ 

lower than that of methane [19], [20]. It should be noted that the lower autoignition temperature of hydrogen leads to limitations in the use of lean premix combustion technology for power plants with a high temperature after compressors. Thus, for engines with air temperature T<sub>k</sub> of the order of 700 K and more, it is necessary to use another combustion system, for example, diffusion or microcluster. However, for engines with T<sub>k</sub> below 700 K, lean burn technology can still be used with high effisiency.



**Figure 7.** - Influence of equivalence ratio on autoignition temperature.

Experimental studies in shock tubes revealed certain dependences of high-temperature ignition on pressure and fuel composition. In [19, 20], three characteristic systems (modes) of ignition were determined depending on the composition of the fuel:

- 1) MCDI (methane chemistry dominating ignition) system for methane-hydrogen fuel mixtures containing less than 40% hydrogen, a decrease in the ignition delay time typical of hydrocarbons with increasing pressure is observed.
- 2) CCMHDI (combined chemistry of methane and hydrogen dominating ignition) system for a 40% CH<sub>4</sub> / 60% H<sub>2</sub> fuel mixture, there is no noticeable effect of pressure on the ignition time. In this case, the nature of ignition is determined neither by the kinetics of hydrogen nor by the kinetics of methane.
- 3) HCDI (hydrogen chemistry dominating ignition) system ignition typical of hydrogen and a complex pressure dependence appear when the mole fraction of hydrogen in the mixture exceeds 80%.

### 1.4. Flame size and form

The flame shape is an important characteristic that affects the temperature field and pollutant emissions [21]. With a strong flow swirl, a typical flow structure is shown in Figure 8. The stream has two recirculation zones: an inner recirculation zone (IRZ) and an outer recirculation zone (ORZ). The fresh air-fuel mixture and combustion products are separated by an inner shear layer (ISL) and an outer shear layer (OSL). The flame is usually V-shaped if the combustion reaction occurs only at the boundary of the internal shear layer (ISL) of the fresh air-fuel mixture and combustion products in the inner recirculation zone (IRZ). If the combustion process occurs at the boundary of the outer shear layer (OSL) of the fresh air-fuel mixture and combustion products in the outer recirculation zone (ORZ), then the flame stabilizes with an M-shape.

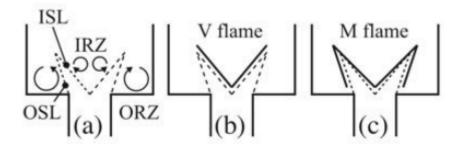


Figure 8. - Scheme of swirling flow in the combustion chamber [21].

In [22] is devoted to an experimental study of the effect of hydrogen enrichment of natural gas on the flame dynamics under atmospheric pressure conditions using a DLE (Dry Low Emission) radially stepped burner. The volume content of hydrogen was 0%, 5%, 11%, 21% and 26%. Authors shows that the shape of the torch changes with an increase in the proportion of hydrogen from M-shaped to V-shaped (Figure 9). Similar results were obtained in [23], where a change in the nature of flame stabilization from an aerodynamically stabilized M-type to stabilization in a V-type shear layer was obtained when the fuel was enriched with hydrogen, and in [24,25], where it was shown that the flame becomes more compact.

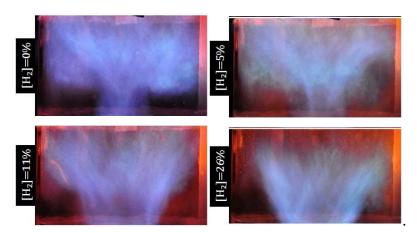


Figure 9. - Change in the shape of the flame with the addition of hydrogen [22].

Similar results were obtained in [26], where it was shown that the flame with a higher proportion of hydrogen is characterized by a more compact shape, which indicates a better reactivity of the fuel mixture (Figure 10).

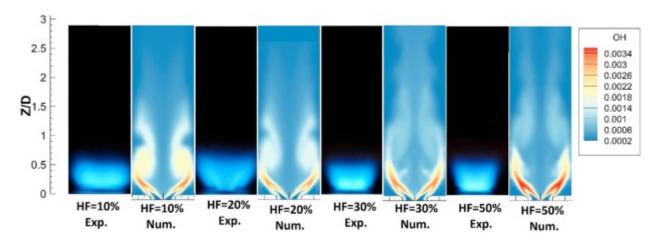
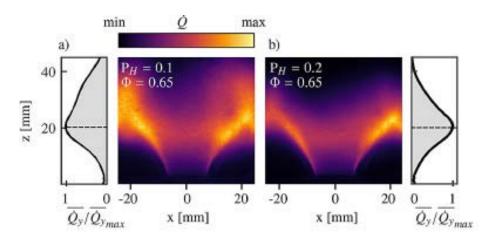


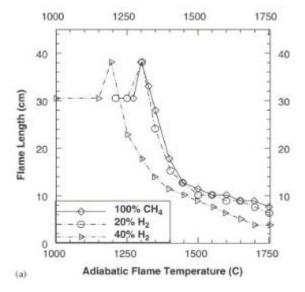
Figure 10. – Flame shame in a swirled flow with different hydrogen addition.

In [27], a premix fuel-air mixture was fed into a cylindrical chamber. The volume concentration range from 0 to 0.5 (where 0 is the absence of hydrogen in the mixture, 1 is pure hydrogen) was investigated, which corresponds to the power fraction  $P_H$  from 0 to 0.25. Flame shapes for two stable operating conditions are shown in figure 11. As can be seen from Figure 11, as the hydrogen content increases, the flame becomes more compact. At  $P_H$ =0.2, the maximum heat release rate remains at the end of the flame brush. It also reduces the interaction with the neighboring flame, as evidenced by the much larger interaction area at  $P_H$ =0.1. For the case of  $P_H$ =0.1, it can be seen that the edges of the torches merge, which creates a large vertically oriented area with a high heat release rate. This is not observed at  $P_H$ =0.2, although there is some evidence of flame tip interaction.



**Figure 11.** - Average flame shapes and corresponding longitudinal distributions of the integral heat release rate for two stable operating conditions [27].

It was found in [28] that the addition of hydrogen at a volume content of less than 20% practically does not affect the flame length, while when using fuel with a hydrogen content of 40% by volume, the flame is much shorter (figure 12).



**Figure 12.** - Dependence of the flame length on the temperature of adiabatic combustion at different volume content of hydrogen [29].

The change in the shape of the flame is associated primarily with an increase in local heat release. As shown in [30], the flame becomes thinner with an increase in the hydrogen

concentration in the methane-hydrogen mixture (Figure 13). This leads to a change in the Karlovitz and Damköhler numbers, as, for example, shown in [31] (Figure 14). Thus, when hydrogen is added to the fuel, the combustion mode of a turbulent flame can be changed from volumetric, when the smallest Kolmagorov turbulent scales penetrate into the flame, to surface, when turbulent vortices only distort, changing its area, but not changing the internal structure. This fact is one of the reasons for the change in the mechanism of the generation of pulsations during the combustion of hydrogen in comparison with the combustion of methane, as will be considered below.

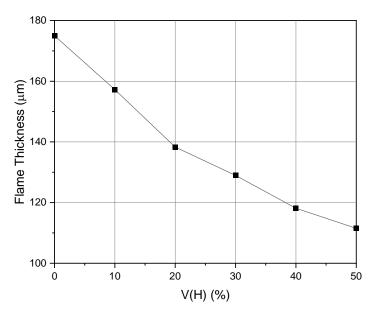


Figure 13. - Flame thickness depending on the proportion of hydrogen in the mixture [30].

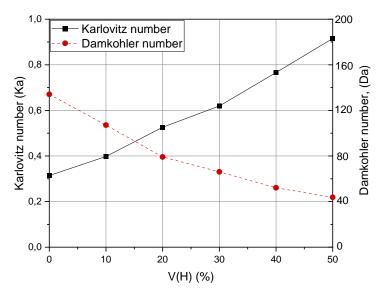


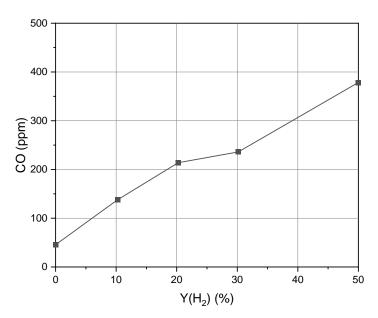
Figure 14. - Karlovitz and Damköhler numbers from hydrogen concentration [30].

It is important to note that the change in these values (Ka and Da) affects the results of calculations of existing mechanisms [32, 33]. Therefore, to predict the operation of engines on hydrogen fuel, new or modified turbulent combustion models should be used, as, for example, shown in [34].

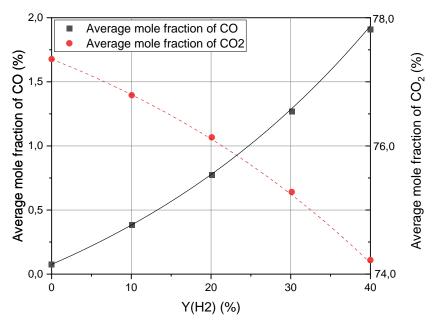
# 2. Influence of features of the behavior of hydrogen combustion processes on the characteristics of combustion chambers

### 2.1. Formation of pollutant emissions: CO and CO2

One of the main benefits of using hydrogen is to reduce the carbon footprint of power plants. Indeed, a decrease in the carbon concentration in the initial fuel leads to an unambiguous decrease in CO<sub>2</sub> in the combustion products per unit of heat received (or per unit mass of fuel). However, when burning methane-hydrogen mixtures, the change in the concentration of CO in the combustion products does not behave so unambiguously. Thus, in [35], [36], an increase in the concentration of CO in combustion products was obtained (Fugure 15, Figure 16).



**Figure 15.** - Dependence of CO emissions on the molar concentration of hydrogen in methane-hydrogen mixture [35], [36].



**Figure 16.** - Dependence of CO and CO2 emissions on the molar concentration of hydrogen in a hydrogen-butane mixture [37].

The increase in CO emissions with increasing hydrogen concentration may be partly due to the shorter residence time of gases in the combustion chamber at higher hydrogen concentrations. Another reason for the increase in CO emissions with hydrogen enrichment is that, due to the higher reactivity and adiabatic flame temperature of hydrogen

compared to hydrocarbon fuels, hydrogen enriched fuel mixtures will burn more efficiently. It is also reported that high temperatures (above the CO<sub>2</sub> dissociation threshold) during combustion of hydrocarbons stimulate CO conversion and therefore lead to more complete combustion and lower CO emissions, but despite all this, competition for oxygen between highly reactive hydrogen and hydrocarbons in the mixture can contribute to an increase in CO emissions [38]. In other words, it can be said that hydrogen inhibits the oxidation of hydrocarbons and their intermediates in an environment with a limited oxygen content (stoichiometric or rich). The competition described above disappears when burning lean mixtures, as shown in Figure 17.

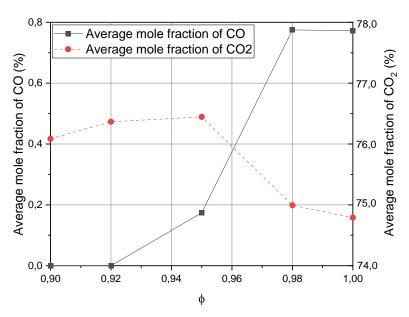


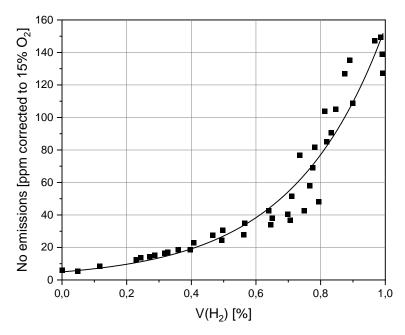
Figure 17. - CO and CO<sub>2</sub> emissions for various equivalence factors [37].

### 2.2. Formation of pollutant emissions: NOx

NOx emissions consist mainly of nitric oxide (NO) and to a lesser extent nitrogen dioxide (NO<sub>2</sub>) and nitrous oxide (N<sub>2</sub>O). NOx is a precursor to photochemical smog, contributes to acid rain and causes ozone depletion. Thus, NOx is a pollutant and is regulated by national and international organizations, in particular ICAO.

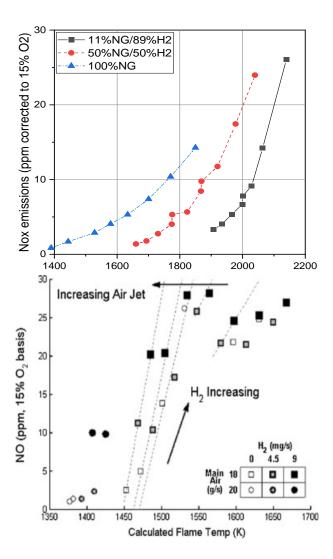
In a laminar flame and at the molecular level in a turbulent flame, the formation of NOx can be explained by four different chemical kinetic processes: thermal NOx, fast (instantaneous) NOx, fuel NOx, and N<sub>2</sub>O intermediate. Thermal NOx are formed as a result of the oxidation of nitrogen present in the atmospheric air supplied for combustion, instantaneous NOx - during high-speed reactions in the flame front, fuel NOx - in the process of oxidation of nitrogen contained in the fuel. Under high pressure and oxygen-rich conditions, NOx can also come from molecular nitrogen (N<sub>2</sub>) via N<sub>2</sub>O.

In [38] [39] present results using various mixtures of natural gas and hydrogen, which show an exponential increase in NO emissions as the percentage of hydrogen increases (Figure 18).



**Figure 18.** - Dependence of NO emissions of natural gas and hydrogen on the volume fraction of hydrogen.

The results in Figure 18 show that there is a significant increase in the rate of NO formation between 60% and 100% hydrogen, which in this work is most likely due to an increase in the rate of hydrogen formation by the thermal mechanism. However, in [4] [40] data are given, normalized to the same temperature in the flame front, which show that at the same combustion temperature, the concentration of nitrogen oxides in the combustion products of methane-hydrogen mixtures is higher than when burning pure methane (Figure 19). This may be due both to a decrease (compactification) of the flame front, which leads to an increase in the residence time of the gas in the high temperature region, and to a change in the chemical mechanism of the formation of nitrogen oxides. Thus, the issue of validating the kinetic mechanisms of the formation of nitrogen oxides during hydrogen combustion is critical, as, for example, in [41].



**Figure 19.** - Dependence of NOx emissions on the adiabatic flame temperature for different proportions of hydrogen-natural gas [4, 41].

# 2.3. Flame blowout and flashback

The reliability of the combustion chamber is determined by such characteristics as lean flame blowout, flashback and pulsating combustion. The addition of hydrogen to methane significantly increases the blowout limits of both suspended and attached diffusion flames [42, 43] (Figure 20). The effect of adding hydrogen to the air flow is much more pronounced than that of adding it to methane (with separate fuel supply). It is shown that the flow velocity on stabilization limits are proportional to the square of the maximum laminar flame propagation velocity. The addition of diluents (CO<sub>2</sub> and N<sub>2</sub>) to the fuel reduced the resistance of the diffusion flame to a much greater extent for the suspended flame than for the attached flame.

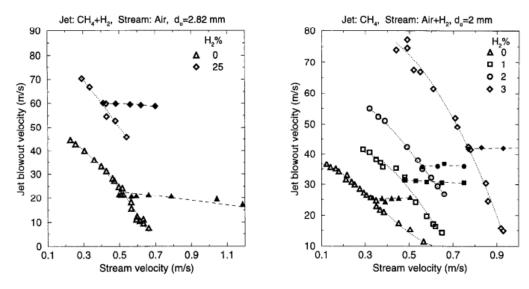


Figure 20. – Flame blowout during combustion of methane and hydrogen methane [42,43].

In [44] (Figure 21) a study was made of the stabilization of the flame of a premixed methane-air mixture with hydrogen additives (up to 29% by mole fraction) behind a bladed swirler. It is shown that the increase in combustion stability upon enrichment with hydrogen is a direct result of higher concentrations of OH, H, and O radicals, which increase several key reaction rates. In this case, the flame extinction time is affected not only by the value of the average OH concentration, but also by its RMS [45]. The greatest effect was achieved with small additions of hydrogen (up to 12–20% [46] by mole fractions), while an increase to 29% by mole fractions, although it had a further positive effect on combustion stability, was not so pronounced.

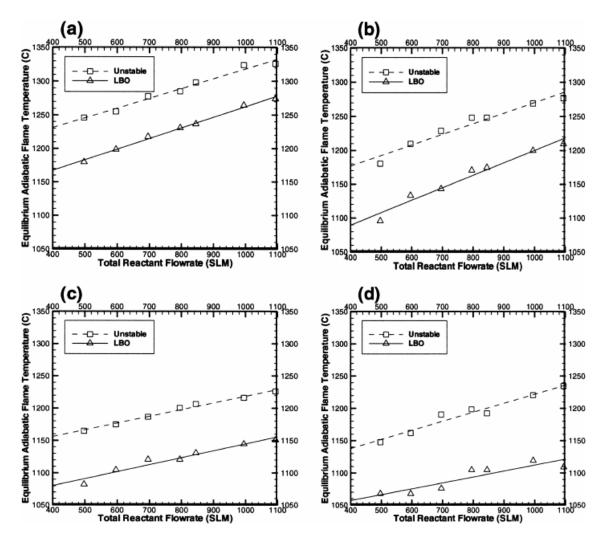
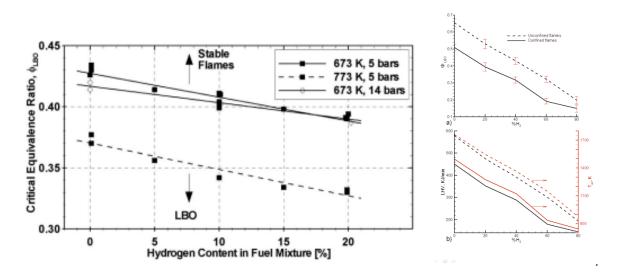


Figure 21. - Change of adiabatic flame temperature during flame blowout.

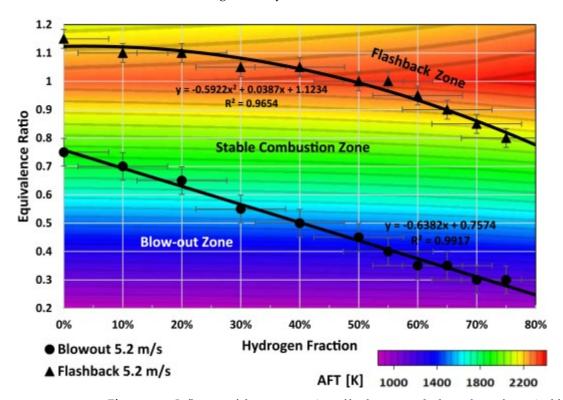
It was shown in [45] (Figure 22) that the addition of hydrogen to methane up to 20% by volume reduces the equivalence ratio during the blowout of a homogeneous flame by 9-10%, and the dependence of  $\Phi$  blowout on the volume content of hydrogen in the fuel is linear, which is also confirmed in [43]. However, in [43], the effect of H<sub>2</sub> on flame blowout is more significant and amounts to about 15–20%  $\Phi$  for every 20% addition of hydrogen.



[45]

Figure 22. - Inluence of Hydrogen content on lean flame blowout.

In [46], a map of the combustion regimes of a methane-hydrogen premix mixture with air was obtained. It was found that with an increase in the proportion of hydrogen, the limits for flame blowout and flashback are shifted to a leaner region, which is associated with an increase in the flame speed and a decrease in the ignition delay time. In this case, the flame can burn at lower temperatures, as shown in Figure 23. Confirmation of this can also be found in [27], where at 40% hydrogen content, the temperature of lean flame blowout is significantly reduced.



**Figure 23.** – Influence of the concentration of hydrogen on the boundary of sustainable combustion for lean flame blowout and flashback [27].

It has also been shown that flame blowout correlates with the Reynolds number to some extent, which is determined by the influence of the intensity of the flow turbulence. In this case, an increase in the proportion of hydrogen in the mixture leads to a decrease in the Reynolds number while maintaining a constant flow rate.

It was shown in [33] that with an increase in the proportion of hydrogen in the fuel, the role of external (corner) recirculation zones in flame stabilization decreases, since the flame is shifted closer to the place of fuel supply. The lean flame blowout limit decreases linearly with the addition of hydrogen, resulting in a stable flame at lower flame temperatures.

When methane was enriched with hydrogen in a ratio of 55%, the flame had the widest stable range (Figure 24). Any attempt to increase the proportion of hydrogen above 55% under stoichiometric conditions (equivalence factor 1.0) failed and the flame went out due to flashback. This is unlike the flame of pure methane, which has proven to be stable under stoichiometric conditions. Increasing the proportion of hydrogen in the fuel mixture increases the laminar flame speed and the flame tries to propagate upstream from the reactant gases, resulting in earlier ignition compared to an oxygen flame of pure methane.

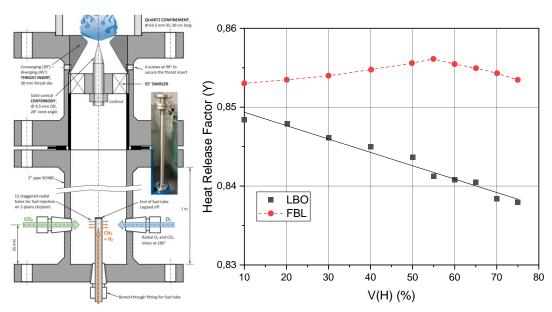
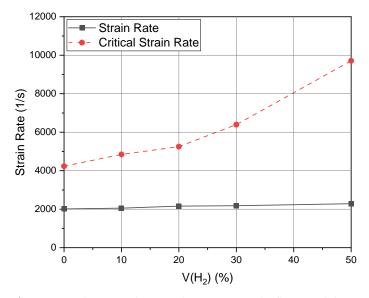


Figure 24. - The range of stable operation of the burner when hydrogen is added to the fuel.

One of the main factors determining the expansion of the combustion limits for lean flame blowout with the addition of hydrogen is an increase in the critical flame deformation rate. Thus, it was shown in [33] that the critical strain rate significantly increases by more than two times from  $\sim$ 4300 s<sup>-1</sup> for the combustion of pure methane to  $\sim$ 9700 s<sup>-1</sup> to expand the boundaries of stable combustion (Figure 25).



**Figure 25.** – Change in the critical strain rate in the flame and the strain rate in the flow with a change in the hydrogen concentration in the fuel [33].

Thus, almost all studies confirm the positive effect of hydrogen additions on the resistance of the lean flame blowout. This also increases the probability of flashback. In general, the range of stable operation shifts to the region of lean mixtures, and the operating window in terms of the excess air coefficient expands, reaching a maximum value at HF $\approx$ 0.55. It is important to note that when hydrogen is added, the working window shifts towards lower flame temperatures, which contributes to a decrease in the rate of formation of nitrogen oxides. Thus, one of the main recommendations when converting combustion chambers to hydrogen is the need to ensure that the temperature in the combustion zone is 100-200 degrees lower (depending on the hydrogen concentration) than when burning methane.

### 2.4. Unsteady combustion

The generation of pulsations in the combustion chamber is often undesirable due to the probability of its complete destruction. At present, the mechanisms of the generation of pulsations during the combustion of natural gas are well described. There are no fundamental changes in the mechanisms of generation during the combustion of hydrogen, however, the characteristics of the flame change, which leads to a change in the mechanisms for the generation of pulsating combustion. The purpose of this section is to analyze the works devoted to the effect of hydrogen additives on unsteady combustion. In this case, several areas of analysis of the effect of hydrogen additives can be distinguished: time-lag, shape and position of the flame, changes in frequency and amplitude, changes in the phase delay.

With an increase in the hydrogen content, the shape, size and location of the flame changes significantly. As shown in [21,22,24,26,33,48-52], the flame becomes more compact (shorter) and pressed against the base of the burner. During combustion of a premix mixture in a swirling flow, the shape of the flame changes from M-shaped to V-shaped. According to [27], such a change in size can occur at a volume percentage of hydrogen of more than 20%. Researches [22] [28] demonstrate that a decrease in the size of flames in an annular combustion chamber leads to a decrease in the probability of their interaction with each other and, consequently, to the elimination of this mechanism for the generation of pulsations. Such an impact on the size and location of the flame is explained by an increase in local heat release and temperature in the combustion zone with an increase in the hydrogen content in the fuel-air mixture. It is also worth noting that the addition of hydrogen leads to a change in the flame stabilization region [24], changing the nature of the flame from suspended to a more stable flame with a fixed stabilization point. In this case, the most stable option would be a fixed point of stabilization of the flame through the use of a pilot circuit or a bluff body with a fixed separation point.

It was shown in [49] that the transition to harmonic oscillations is preceded by the appearance of a flame in the outer recirculation zone, which, for the combustion of methane–hydrogen mixtures, occurs at leaner fuel–air mixture compositions than during combustion of methane. This transition can be estimated using the function of the dimensionless strain rate (normalized strain rate):  $\frac{k_{ext}D}{U_{\infty}}$ .. In [23], a "center of heat release" was proposed as an indicator of stability. Because a flame having the same "heat center" location but different operating conditions and fuel composition has almost the same shape.

In [22], the effect of adding hydrogen to a lean mixture of natural gas with air during combustion of a premixed mixture in a swirling flow is studied. The proportion of hydrogen in the fuel-air mixture varied up to 50% by volume. As a result of the work, the influence of the addition of hydrogen on the shape of the flame, the characteristics of the structure of the precessing vortex core (PVC), as well as the frequency and amplitude of pulsations were investigated. Regarding the thermoacoustic characteristics, it was found that with an increase in the proportion of hydrogen in the fuel (as well as with an increase in the air temperature), the speed of sound between the fuel supply point and the flame front (in the premixer) increases due to a decrease in the average molecular weight of the mixture with the addition of hydrogen (Figure 26). This characteristic will reflect the time of transfer of acoustic energy from the flame front, as a source, to the place of fuel supply, as a factor that further leads to an increase in pulsations. Thus, both the addition of hydrogen and a decrease in the flame size lead to a decrease in the time of acoustic energy transfer from the flame front to the place of fuel supply, affecting the pulsation frequency and increasing it [22]. In this case, the pulsation frequency with an M-type flame is lower than with a V-type.

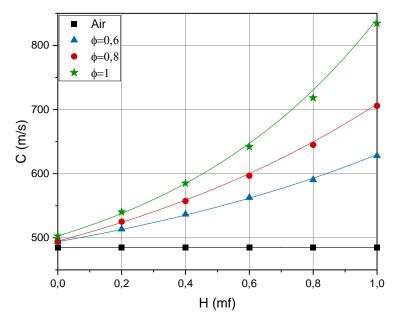


Figure 26. - Influence of the molar fraction of hydrogen on the speed of sound.

Similar conclusions were obtained in [47] devoted to the study of the effect of hydrogen addition to the methane-air mixture on the self-excited thermoacoustic instability of the flame, including when the length of the combustion chamber was changed in the range from 300 to 1100 mm. With an increase in the proportion of hydrogen, the size of the flame also decreases (Figure 1.30). At a low hydrogen concentration (~15%), low-frequency primary acoustic modes (<200 Hz) were mainly excited. When the frequency of the primary oscillations (without addition of hydrogen) was less than 200 Hz, the frequency tended to increase linearly with the percentage of hydrogen. However, a linear relationship between frequency and hydrogen fraction has been associated with acoustic modes that are influenced by chamber length. At high hydrogen concentrations (>40%), primary acoustic modes tended to occur at higher frequencies (~400 Hz). Increasing the hydrogen content leads to a decrease in flame length (Figure 27).

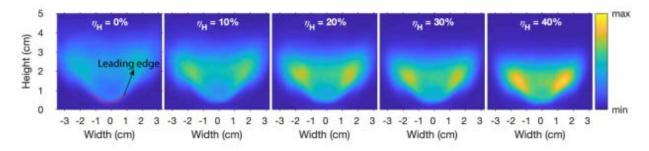


Figure 27. - Dependence of the flame length on the hydrogen content [47].

In [48, 49], the observed frequencies coincided with the frequencies of the longitudinal modes of the chamber, while in [48] the hydrogen content did not affect the pulsation frequency, which remained equal to the frequency of the first longitudinal mode of the combustion chamber. On the other hand, the authors of [22, 47] noted an increase in the frequencies of the first modes with an increase in the hydrogen content. This is due to an increase in the temperature of the combustion products and, as a consequence, an increase in the speed of sound in the combustion chamber. While maintaining the temperature of the combustion products, the pulsation frequency can remain the same.

It was shown in [27] that with an increase in the fraction of the hydrogen content, the acoustic energy is redistributed between frequencies. Thus, when burning pure methane,

the authors identified three main frequencies of 275 Hz, 450 Hz, 600 Hz, while when burning 60% of hydrogen, one frequency is characteristic - 450 Hz. The absence of the 600 Hz band indicates a change in the acoustic damping/amplification characteristics (amplitudes) of the combustion chamber, providing a high intensity in the 450 Hz band. This mode of combustion is more dangerous, since it can lead to fatigue failure of the elements of the combustion chamber.

It was shown in [49] that upon transition to the oscillation mode, the pulsation frequency during the combustion of a hydrogen–methane mixture is equal to the pulsation frequency during the combustion of pure methane (110 Hz). However, at an excess air ratio of 0.65, the instability in the combustion of the hydrogen-methane mixture jumped to a higher frequency of 180 Hz. All observed frequencies (110, 180 Hz) coincide with the frequencies of the longitudinal modes of the combustion chamber.

The influence of the hydrogen content on the amplitude of pressure fluctuations depends on the design and operating parameters of the combustion chamber: the length of the combustion chamber, average pressure, excess air coefficient, acoustic conditions at the inlet and outlet of the chamber, swirl number, etc.

It was noted in [47] that a low hydrogen content (~15%) can enhance pulsations, while a sufficiently high hydrogen level (~40%) can weaken the primary modes. In turn, in [26], the largest amplitude was found at 25% hydrogen content in the fuel by volume and decreased with a further increase in the hydrogen content. In [27], when studying the combustion of a premixed mixture in a swirling flow, it was found that the noise level is practically independent of the volumetric content of hydrogen, with the exception of a fuel-air mixture with a hydrogen content of 40% at an adiabatic temperature Tad> 1500°C. These results indicate a transition from a stable combustion regime to an unstable one with an increase in the hydrogen volume concentration. In [21], a decrease in the amplitude of pulsations is observed in the entire range of hydrogen content studied (5-26% by volume). The explanation of the multidirectional influence of the addition of hydrogen can be related to the initial phase delay between heat release pulsations and pressure pulsations (Figure 28). If during the combustion of methane the phase delay was negative, that is, pressure pulsations occur after heat release pulsations (at a phase angle of  $0>\varphi>90$ ), the addition of hydrogen increases the amplitude of pulsations. If in the initial configuration without hydrogen the phase delay was positive, that is, pressure pulsations occurred before heat release pulsations (at a phase angle of  $0>\varphi>90$ ), the addition of hydrogen reduces the pulsation amplitude. If, in the initial configuration, the acoustic pressure and changes in heat release were in phase, any change in the hydrogen content reduces the amplitude of the pulsations. That is, the frequency of pulsations, as a rule, increases.

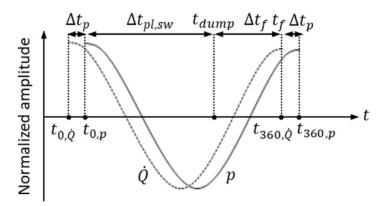


Figure 28. - Scheme of the amplitude of pressure fluctuations in the fuel-air mixture flame.

With an increase in the hydrogen content, the frequency distribution of the power spectral density can change [27]. At the same time, significant changes in the amplitude of pulsations occur when hydrogen is added to the fuel from 30-40% and higher in volume.

The authors come to the conclusion that the dynamic stability of the combustion chamber they analyzed with a change in the composition of the fuel and operating conditions was largely determined by the position of the center of the heat release flame (the middle location of the flame) under stable conditions [23].

### 3. Conclusion

As a result of the analysis of the literature on the study of hydrogen combustion, the following conclusions and recommendations can be drawn:

- The density of hydrogen is almost an order of magnitude lower than the density of natural gas, and the net calorific value per cubic meter is several times lower. Thus, while maintaining the parameters of the power plant when switching to hydrogen, it is necessary to pay attention to the flow sections of the fuel supply system, especially fuel injectors.
- The addition of hydrogen leads to an increase in the intensity of chemical processes per unit volume in the flame front. This leads to the compactification of the flame, a change in its shape and a decrease in its size.
- The adiabatic combustion temperature of a hydrogen flame is 5-15% higher than that of methane, however, the operating range of hydrogen covers lower minimum temperatures up to 1050 K, compared to ~1400-1500 K for methane, due to the high concentration of active radicals (H, HO).
- The addition of hydrogen results in a significant increase in flame propagation speed. The greatest effect is achieved at a hydrogen concentration in the fuel of 65% or more. In this case, the maximum velocity shifts to the region of rich mixtures. Based on the high hydrogen velocity, the combustion chamber can theoretically be shortened, which will reduce both engine weight and NOx emissions by reducing the residence time of the air-fuel mixture in high temperature zones.
- To ensure combustion without flashback, it is necessary to increase the flow rate, which requires an increase in the pressure drop across the flame tube head and, in general, an increase in pressure losses in the combustion chamber.
- The ignition delay time of hydrogen is several orders of magnitude shorter than that of methane. The minimum autoignition temperature shifts to the region of rich mixtures and is approximately 70-150 degrees lower than that of methane. Thus, for combustion chambers with air temperature Tk>700 K, it is advisable to use a combustion system without premixing, for example, LDI or multicluster.
- The emission of CO increases when hydrogen is added to the fuel, while CO<sub>2</sub> decreases for stoichiometric and rich mixtures. When burning lean mixtures of hydrogen with air, it is possible to achieve a reduction in the level of CO and CO<sub>2</sub> emissions compared to the combustion of methane.
- The emission of NOx when hydrogen is added to the fuel increases at the same adibatic temperature of the flame, which indicates different mechanisms for the formation of nitrogen oxides during the combustion of methane and metahydrogen mixtures. However, nitrogen oxide emissions can be achieved by lowering the combustion temperature.
- An increase in the concentration of hydrogen in mixtures with methane and natural gas is accompanied by an increase in resistance to flame blowout, but increases the tendency of the mixture to flashback [52].
- The change in the amplitude of pulsations with an increase in the concentration of hydrogen in the fuel will be determined by the initial mechanism of their formation. In this case, the frequency of pulsations, as a rule, increases.
- In general, the effect of increased pressure on a hydrogen-air flame is more pronounced than for a methane-air flame, which complicates the process of interpolating data obtained under atmospheric conditions for motor parameters. Thus, the issue of further research into the influence of pressure on the combustion of hydrogen is topical.

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#### References

- 1. Russian Government. Avilable online: http://government.ru/docs/42971/
- 2. Acar, C.; Dincer, I. The potential role of hydrogen as a sustainable transportation fuel to combat global warming. International Journal of Hydrogen Energy **2020**, 45, 3396-3406.
- 3. Taamallah, S.; Vogiatzaki, K.; Alzahrani, F.M.; Mokheimer, E.M.A.; Habib, M.A.; Ghoniem, A.F. Fuel flexibility, stability and emissions in premixed hydrogen-rich gas turbine combustion: Technology, fundamentals, and numerical simulations. Applied Energy 2015, 154, 1020-1047.
- Griebel, P. Gas Turbines and Hydrogen. Hydrogen Science and Engineering: Materials, Processes, Systems and Technology 2016, 2, 1011-1032.
- 5. Biryuk, V.; Lukachev, S.; Uglanov, D.; Tsybizov, Y. Gas in engines, 1st ed.; Publishing house of Samara University: Samara, Russia, 2021; pp. 3-296.
- Wallner, T.; Ng, H.K.; Peters, R.W. The effects of blending hydrogen with methane on engine operation, efficiency, and emissions. SAE Technical Papers 2007.
- Wei, J. Boiling points and melting points of chlorofluprocarbons. Industrial and Engineering Chemistry Research 2000, 39, 3116-3119.
- 8. GRI-Mech 3.0. Avilable online: http://combustion.berkeley.edu/gri-mech/version30/text30.html
- 9. Boushaki, T.; Dhué, Y.; Selle, L.; Ferret, B.; Poinsot, T. Effects of hydrogen and steam addition on laminar burning velocity of methane-air premixed flame: Experimental and numerical analysis. International Journal of Hydrogen Energy 2012, 37, 9412-9422.
- 10. Sun, Y.; Zhang, Y.; Huang, M.; Li, Q.; Wang, W.; Zhao, D.; Cheng, S.; Deng, H.; Du, J.; Song, Y.; Li, H.; Xu, H. Effect of hydrogen addition on the combustion and emission characteristics of methane under gas turbine relevant operating condition. Fuel **2022**, 324.
- 11. Li, Z.; Cheng, X.; Wei, W.; Qiu, L.; Wu, H. Effects of hydrogen addition on laminar flame speeds of methane, ethane and propane: Experimental and numerical analysis. International Journal of Hydrogen Energy **2017**, 42, 24055-24066.
- 12. Ilbas, M.; Crayford, A.P.; Yilmaz, I.; Bowen, P.J.; Syred, N. Laminar-burning velocities of hydrogen-air and hydrogen-methane-air mixtures: An experimental study. International Journal of Hydrogen Energy **2006**, 31, 1768-1779.
- Zhang, Y.; Huang, Z.; Wei, L.; Zhang, J.; Law, C.K. Experimental and modeling study on ignition delays of lean mixtures of methane, hydrogen, oxygen, and argon at elevated pressures. Combustion and Flame 2012, 159, 918-931.
- 14. Herzler, J.; Naumann, C. Shock-tube study of the ignition of methane/ethane/hydrogen mixtures with hydrogen contents from 0% to 100% at different pressures. Proceedings of the Combustion Institute **2009**, 32 I, 212-220.
- 15. Lieuwen, T.; McDonell, V.; Petersen, E.; Santavicca, D. Fuel flexibility influences on premixed combustor blowout, flashback, autoignition, and stability. Journal of Engineering for Gas Turbines and Power 2008, 130, 1.
- Beerer D.J. Autoignition of methane, ethane, propane and hydrogen in turbulent high pressure and temperature flows. Master's thesis, The University of California, Irvine, California, USA; 2009.
- 17. Explosionprotected electrical apparatus. Part 4. Method of test for ignition temperature. Avilable online: <a href="https://docs.cntd.ru/document/1200103115?ysclid=lbeo3uxkcl682084142">https://docs.cntd.ru/document/1200103115?ysclid=lbeo3uxkcl682084142</a>
- 18. Zhang, Y.; Jiang, X.; Wei, L.; Zhang, J.; Tang, C.; Huang Z. Experimental and modeling study on auto-ignition characteristics of methane/hydrogen blends under engine relevant pressure. International Journal of Hydrogen Energy **2012**, 37, 19168-19176.
- 19. Zhang, Y.; Huang, Z.; Experimental and modeling study on auto-ignition of methane/hydrogen blends at elevated pressures. SAE Technical Papers **2014**, 1.
- 20. Gupta, A.K.; Lilley, D.G.; Syred N. Swirl flows. Combustion and Flame 1984, 63, 311-311.
- 21. Ge, B.; Ji, Y.; Zhang, Z.; Zang, S.; Tian, Y.; Yu, H.; Chen, M.; Jiao, G.; Zhang, D. Experiment study on the combustion performance of hydrogen-enriched natural gas in a DLE burner. International Journal of Hydrogen Energy **2019**, 44, 14023-14031.
- 22. Chterev, I. Boxx, I. Effect of hydrogen enrichment on the dynamics of a lean technically premixed elevated pressure flame. Combustion and Flame **2021**, 225, 149-159.
- 23. Figura, L.; Lee, J.G.; Quay, B.D.; Santavicca, D.A. The Effects of Fuel Composition on Flame Structure and Combustion Dynamics in a Lean Premixed Combustor. ASME **2007**, *2*, 181-187.
- 24. Yilmaz, H.; Cam, O.; Yilmaz, I. Experimental investigation of flame instability in a premixed combustor. Fuel 2020, 262.
- 25. Hu, Z.; Zhang, X. Experimental study on flame stability of biogas / hydrogen combustion. International Journal of Hydrogen Energy **2019**, 5607-5614.
- 26. Indlekofer, T.; Ahn, B.; Kwah, Y.H.; Wiseman, S.; Mazur, M.; Dawson, J.R.; Worth, N.A. The effect of hydrogen addition on the amplitude and harmonic response of azimuthal instabilities in a pressurized annular combustor. Combustion and Flame 2021, 228, 375-387.
- 27. Wicksall, D.M.; Agrawal, A.K. Acoustics measurements in a lean premixed combustor operated on hydrogen/hydrocarbon fuel mixtures. International Journal of Hydrogen Energy 2007, 32, 1103-1112.
- 28. Kashir, B.; Tabejamaat, S. A numerical study on the effects of H2 addition in non-premixed turbulent combustion of C3H8-H2-N2 mixture using a steady flamelet approach. International Journal of Hydrogen Energy **2013**, 38, 9918-9927.
- 29. Araoye, A.A.; Abdelhafez, A.; Nemitallah, M.A.; Habib, M.A.; Ben-Mansour, R. Experimental and numerical investigation of stability and emissions of hydrogen-assisted oxy-methane flames in a multi-hole model gas-turbine burner. International Journal of Hydrogen Energy **2021**, 46, 20093-20106.

- Bongartz, D.; Ghoniem, A.F. Chemical kinetics mechanism for oxy-fuel combustion of mixtures of hydrogen sulfide and methane. Combustion and Flame 2015, 162, 544-553.
- 31. Zhang, P.; Zsély, I.G.; Papp, M.; Nagy, T.; Turányi, T. Comparison of methane combustion mechanisms using laminar burning velocity measurements. Combustion and Flame **2022**, 238.
- 32. Verma, S.; Monnier, F.; Lipatnikov, A.N. Validation of leading point concept in RANS simulations of highly turbulent lean syngas-air flames with well-pronounced diffusional-thermal effects. International Journal of Hydrogen Energy 2021, 46, 9222-9233
- 33. Nemitallah, M.A.; Imteyaz, B.; Abdelhafez, A.; Habib, M.A. Experimental and computational study on stability characteristics of hydrogen-enriched oxy-methane premixed flames. Applied Energy **2019**, 250, 433-443.
- 34. Bouras, F.; El Hadi Attia, M.; Khaldi, F.; Si-Ameur, M. Control of methane flame properties by hydrogen fuel addition: Application to power plant combustion chamber. International Journal of Hydrogen Energy **2017**, 42, 8932-8939.
- 35. Abubakar, Z.; Shakeel, M.R.; Mokheimer, E.M.A. Experimental and numerical analysis of non-premixed oxy-combustion of hydrogen-enriched propane in a swirl stabilized combustor. Energy **2018**, 165, 1401-1414.
- 36. Therkelsen, P.; Werts, T.; McDonell, V.; Samuelsen, S. Analysis of NOx Formation in a Hydrogen-Fueled Gas Turbine Engine. ASME 2009, 131, 031507.
- 37. Hashemi, S.A.; Hajialigol, N.; Mazaheri, K.; Fattahi, A. Investigation of the effect of the flame holder geometry on the flame structure in non-premixed hydrogen-hydrocarbon composite fuel combustion. Combustion, Explosion and Shock Waves 2014, 50, 32-41.
- 38. Ghoniem, A.F.; Annaswamy, A.; Park, S.; Sobhani, Z.C. Stability and emissions control using air injection and H2 addition in premixed combustion. Proceedings of the Combustion Institute **2005**, 30 II, 1765-1773.
- 39. Semenikhin, A.S.; Matveev, S.S.; Chechet, I.V.; Matveev, S.G.; Idrisov, D.V.; Gurakov, N.I.; Radin, D.V.; Novichkova, S.S.; Fokin, N.I.; Simin, N.O.; Ivanovskii, A.A.; Tarasov, D.S. Kinetic Models of Methane-Hydrogen Mixture Combustion: Brief Review and Validation. Thermal Engineering 2022, 69, 792-801.
- 40. Papanikolaou, N.; Wierzba, I. The effects of burner geometry and fuel composition on the stability of a jet diffusion flame. Journal of Energy Resources Technology, Transactions of the ASME **1997**, 119, 265-270.
- 41. Karbasi, M.; Wierzba, I. The effects of hydrogen addition on the stability limits of methane jet diffusion flames. International Journal of Hydrogen Energy **1998**, 23, 123-129.
- 42. Schefer, R.W. Wicksall, D.M. Agrawal, A.K. Combustion of hydrogen-enriched methane in a lean premixed swirl-stabilized burner. Proceedings of the Combustion Institute **2002**, 29, 843-851.
- Zhu, S.; Acharya, S. An experimental study of lean blowout with hydrogen-enriched fuels. Journal of Engineering for Gas Turbines and Power 2012, 134.
- Schefer, R.W. Hydrogen enrichment for improved lean flame stability. International Journal of Hydrogen Energy 2003, 28, 1131-1141.
- 45. Griebel, P.; Boschek, E.; Jansohn, P. Lean Blowout Limits and NOx Emissions of Turbulent, Lean Premixed, Hydrogen-Enriched Methane/Air Flames at High Pressure. ASME **2006**, 1, 405-412.
- 46. Imteyaz, B.A.; Nemitallah, M.A.; Abdelhafez, A.A.; Habib, M.A. Combustion behavior and stability map of hydrogen-enriched oxy-methane premixed flames in a model gas turbine combustor. International Journal of Hydrogen Energy 2018, 43, 16652-16666.
- 47. Zhang, J.; Ratner, A. Experimental study of the effects of hydrogen addition on the thermoacoustic instability in a variable-length combustor. International Journal of Hydrogen Energy **2021**, 46, 16086-16100.
- 48. Davis, D.W.; Therkelsen, P.L.; Littlejohn, D.; Cheng, R.K. Effects of hydrogen on the thermo-acoustics coupling mechanisms of low-swirl injector flames in a model gas turbine combustor. Proceedings of the Combustion Institute **2013**, 34, 3135-3143.
- 49. Shanbhogue, S.J.; Sanusi, Y.S.; Taamallah, S.; Habib, M.A.; Mokheimer, E.M.A. Ghoniem, A. F. Flame macrostructures, combustion instability and extinction strain scaling in swirl-stabilized premixed CH4/H2 combustion. Combustion and Flame **2016**, 167, 497.
- 50. Zhang, J.; Ratner, A. Experimental study on the excitation of thermoacoustic instability of hydrogen-methane/air premixed flames under atmospheric and elevated pressure conditions. International Journal of Hydrogen Energy **2019**, 44, 21324-21335.
- 51. Yilmaz, I.; Ratner, A.; Ilbas, M.; Huang, Y. Experimental investigation of thermoacoustic coupling using blended hydrogenmethane fuels in a low swirl burner. International Journal of Hydrogen Energy **2010**, 35, 329-336.
- 52. El-Ghafour, S.A.A.; El-dein, A.H.E.; Aref, A.A.R. Combustion characteristics of natural gas-hydrogen hybrid fuel turbulent diffusion flame. International Journal of Hydrogen Energy **2010**, 35, 2556-2565.