

Renewable hydrogen in gas grids, Effects on laminar burning velocities

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Abstract—Hydrogen can be injected into the gas grid by using different power to gas methods, so the combustion influence of hydrogen is more and more of interest. Furthermore, the laminar burning velocity is one fundamental property of a reactive oxidizer-fuel mixtures, varying with pressure, initial composition and temperature. These values are important for validation of reaction mechanisms and the specific design of industrial burners. There are several experimental methods to measure laminar burning velocities, e.g. the stagnation flame method, the Bunsen flame method, the spherically expanding flame method and the flat flame burner method, which also includes the heat-flux burner method. The accuracy of the different methods could be strongly enhanced over the last ten years but there are still uncertainties of up to 25 % depending on method, boundary condition and fuel composition. Furthermore, for a lot of fuels, especially hydrogen containing fuel blends, there is a lack of experimental data. In this study, the heat-flux burner method was applied to measure the laminar burning velocities of hydrogen containing fuel blends. These fuel blends are of major interest since the hydrogen concentration in the gas grid system could in future rise to a significant value. For example, within the “power-to-gas” concept, some new technologies arise to use renewable energy to produce hydrogen and take it as “fuel”. So the change of the gas composition also changes the combustion properties, for example the burning velocity, heating value and ignition delay. Therefore, different hydrogen containing fuels were tested within a wide range of equivalence ratios between 0.7 and 1.6. Initial temperatures of 293 K up to 383 K were tested for atmospheric conditions. With these results the lack of data for hydrogen containing gases could be significantly closed and helps to validate reaction mechanisms for better simulation of internal combustion systems.

I. INTRODUCTION

Since the beginning of industrialization, the global consumption of fossil fuels has been steadily increased. These resources are limited, so energy must be provided in a different way in the future. For this reason, it is important to use existing resources in an environmentally friendly and effective manner. For a sustainable use of the reserves, low-emission combustion systems with a high efficiency are a key technology to fulfil these requirements. Combustion systems based on gaseous fuels are more environmentally friendly compared to oil and coal fired combustion plants due to the more favorable carbon-to-hydrogen ratio, fewer combustion products and resulting low carbon dioxide emissions per unit of energy.

An increased amount of gaseous fuels from non-conventional sources, such as bio-methane, biogases and hydrogen, is being injected into the German and European gas grids. This has led to a wide scattering of gas composition. Non-conventional natural gas represents 63% of the total natural gas resources [1]. Furthermore, Hydrogen can be injected into the gas grid by using different power to gas methods, so the combustion influence of hydrogen is more and more of interest. Combustion in engines directly is only one attractive way to use hydrogen as an energy carrier. Moreover, it becomes more important in several applications, due to energy efficiency and emission aspects in industrial applications. On the other side the European parliament provide a framework for setting requirements for the environmentally design of energy-related products with the new designed 2009/125/EG [2]. For these new thermal process technologies, the specific information of physic-chemical properties of these fuels are limited and classical knowledge of combustion of small-hydrocarbons (C_1 to C_4) cannot be transferred directly [3].

For these reasons, the associated chemical kinetics and transport properties for the combustion of hydrogen have been studied extensively. Experimental tests with different parameters for various kinds of combustion processes are important in order to distinguish between the different predictions and determine areas of uncertainty, possibly ultimately leading to improved parameter values. Therefore, new experimental results are reported here that can be used to test predictions of methane and methane-hydrogen mixtures combustion.

The laminar burning velocity of an adiabatic laminar flame s_L is a key chemical data for combustion processes as well as combustion models. The laminar burning velocity is also a fundamental property of reactive fuel-oxidizer mixtures, varying with composition, pressure and initial temperature and is very important for validation of reaction mechanisms and the specific design of burners for thermal industry and household applications.

II. EXPERIMENTAL METHOD

A. Definitions and principle of the method

Laminar burning velocity is the speed of a free-propagating planar adiabatic flame relative to the unburned mixture. While direct implementation of the s_L definition for its measurement is not possible, since it requires to create an infinite and perfectly planar flame. [4] Nowadays several approaches are used to measure the laminar burning velocity. Each of them was built on

different physical principles and has his own advantage and range of applicability. Mostly counter-flow burner [5], spherical expanding flame chambers [6], flat-flame burner [7] as well as the heat-flux burner [8] are in use. The latter is superior to a flat flame burner due to the realization of a quasi-adiabatic flame by compensation of the heat loss of the flame. Furthermore, additionally no Markstein correction for the normal velocity vector has to be applied [9]. Due to the quasi-adiabatic compensation and nearly up-stretched flame shape, this type of burner is currently seen to be among the most accurate methods to determine laminar burning velocities in atmospheric conditions. Gaswani et al. [10] reached measurements of syngas up to 10atm for the heat-flux method. The heat-flux method also has a limitation in velocities, so that S_L up to about 60 cm/s can be measured. Above that limit the area would become disturbed by the perforated heated burner plate. [4] The main advantage of the heat flux burner is that the flames are flat in the area of measurements. Therefore, a direct determination of the laminar burning velocity is possible without correcting the stretch and strain with mathematical extrapolation. This avoids an additional uncertainty made with other methods. In this work, we used such a home-build heat-flux burner to measure adiabatic laminar burning velocities of gaseous fuels within a range of equivalence ratios from 0.7 to 1.4 and initial temperatures of 298 K up to 363 K in atmospheric conditions under 60 cm/s.

B. Heat-flux burner setup at TU Bergakademie Freiberg

The used heat-flux burner was proposed by de Goey, van Maaren and Quax in 1993 [7] and is based on experimental work of Botha and Spalding in 1954 [11]. The advantage of this method for measuring the laminar burning velocity is the investigation of an unstretched flame under quasi adiabatic conditions. The stabilization of a planar flame has been further proved by van Maaren et al. [12]. The heat-flux burner which was used at the TUBAF (TU Bergakademie Freiberg) is shown in Fig. 2. It consists of a 2 mm thick brass burner plate with a diameter of 30 mm and a uniform perforation pattern of holes with diameters of 0.5mm. The heat-flux burner plate was designed for burning velocities between around 10 cm/s and 60 cm/s and is surrounded by a heating jacket which can be operated in the temperature region between 273 K and 383 K. In the following experiments the heating jacket operates with a constant temperature of 298 K and 363 K, respectively. The Plenum chamber on the other side was surrounded by a cooling jacket, which regulated the temperature of the plenum chamber to 298 K. In order to measure the radial temperature profile of the burner plate seven thermocouples of type E were attached to the burner plate in distances of 12.6 mm, 9.8 mm, 6.3 mm, 4.2 mm, 2.1 mm and 0 mm to the centre of the plate. The measured temperature profile allows the determination of the net heat flux which results from the sum of the heat flux from the flame front to the burner plate and the heat flux from the burner plate to the premixed fuel-air mixture. If the velocity of the gas u_g was lower than the laminar burning velocity S_L a net heat flux with a positive value occurs leading to a burner plate with a higher temperature than the heating jacket. In this case a heat flux from the centre to the outer ring of the burner plate emerges. Vice versa, a negative net heat flux indicates higher gas velocity compared to the laminar burning velocity. For this condition an inverse heat flux and a centre with the lowest measured temperature follows. Only for a constant temperature profile of all thermocouples both velocities coincide $u_g = S_L$ due to a vanishing heat flux. Therefore, this method offers the possibility to extrapolate the quasi adiabatic state from multiple measurements with $u_g < S_L$ and $u_g > S_L$, whereas for the latter case commonly only one measurement is sufficient.

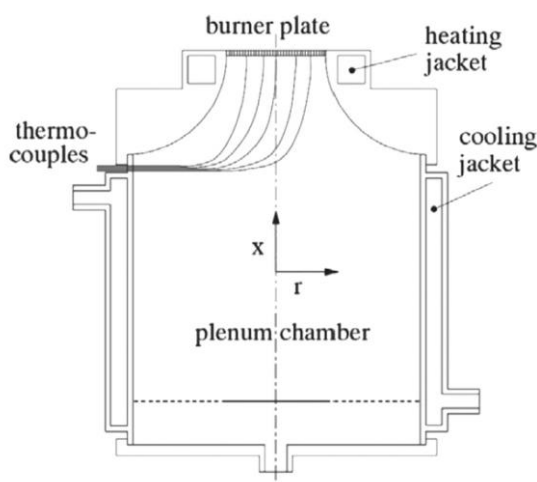


Fig. 1 Schematic view of the utilized heat-Flux burner [10]

At the TU Bergakademie Freiberg setup the gas flows were controlled by five mass flow controllers (MFC) with an uncertainty of 0.8% of the reading value and 0.2 % of the full scale value. These gases were mixed with air in a specially designed and heated chamber in order to get a homogenous gas mixture. The air flow was regulated by a MFC as well, depending on the equivalence ratio. Before each measurement the MFC setup was calibrated with a calibration system of type Definer 220, Bios Corp [13] which corrects the volumetric flow for temperature and pressure. The whole setup is shown in a flowchart of the heat-

flux burner test rig in Fig. 2. The heat-flux burner test rig was automated by a program based on LabVIEW™ from National Instruments Corp. and gives the possibility to control air and gas flows. Furthermore, it logs the ambient conditions as well as the signals from the thermocouples and executes the post processing and subsequent computation of the laminar burning velocities [14].

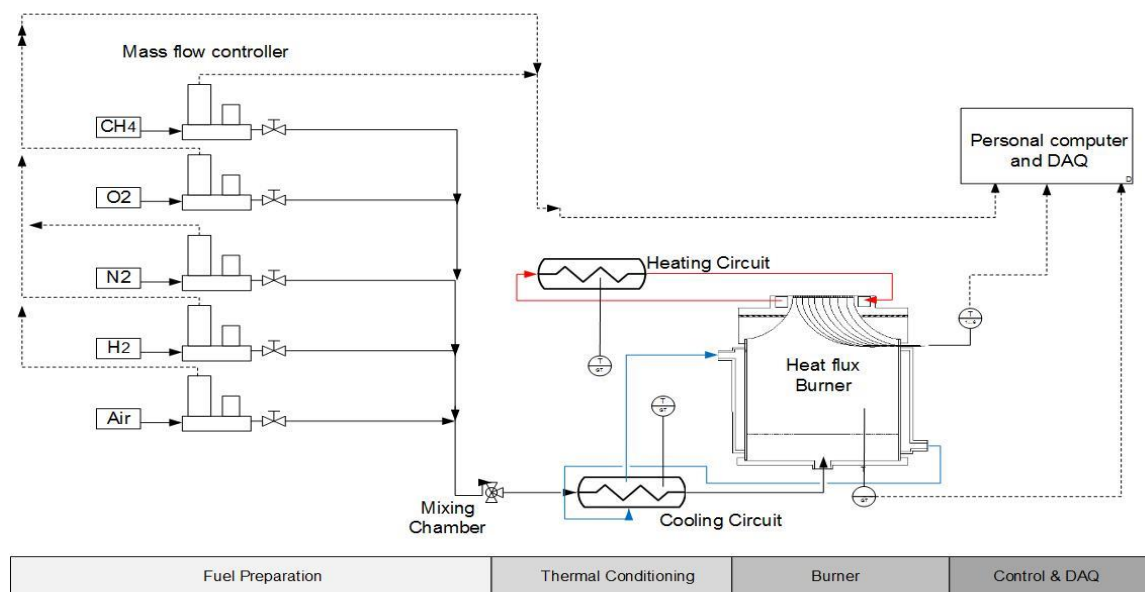


Fig. 2 Flowchart of the heat-flux burner test rig structured in four sections [12]

III. NUMERICAL APPROACH

The Chemkin® software package was used to simulate the combustion process of the heat-flux burner with the “Premixed Laminar Burner-stabilized Flame” model. For the simulations, more than 1150 grid points were used for the computational domain of a spatial dimension of 10 cm to ensure chemical equilibrium in the burned gas region. The grid independence was proved. During this work, four different reaction mechanism were adopted, namely Davis- [15], Li- [16], USC II- [17] and Gri-Mech 3.0 [18] mechanism. For the numerical calculation of the laminar burning velocity of methane, the reaction mechanism Gri-Mech 3.0 was utilized. The reaction mechanism of Davis et al. (2005) was used for simulating the combustion of hydrogen. This mechanism includes 38 elementary reactions and 14 species. The Davis mechanism is optimized for H₂-CO reactions at high temperatures and extensively validated with experimental work [15]. The Li-mechanism, which was developed to describe the H₂-O₂ systems, contains 15 species and 45 elementary reactions. The mechanism was developed for a wide range of process conditions, but is especially validated for high pressures. The USC-II mechanism was primarily designed for the burning of short-chain hydrocarbons (C₁-C₄) and synthesis gas mixtures (H₂/CO). This includes 15 species and 48 reactions; this reaction model is also validated for extensive process conditions. In Table 1 the adopted mechanisms are show in comparison.

TABLE 1 OVERVIEW of UTILIZED REACTION MECHANISM

Reaction mechanism	Number of species	Number of elementary reactions	Year of publication
Gri-Mech 3.0	53	325	2000
Davis	14	38	2005
Li	15	45	2007
USC II	14	48	2007

IV. Results and discussion

A. Laminar burning velocity of methane/air flames and influence of inert gases

Several different gaseous mixtures with different fuel compositions were investigated and are presented in Table 2. The focus of this work has been placed on different groups of fuel composition, namely CH₄-H₂, H₂-N₂, CH₄-N₂ and CH₄-CO₂. The following cases are inspired from German gas grid composition [19] and typical gas compositions for different industrial applications. When these gases are encouraged, large fluctuations occur in the flow rate and the concentration of the gas

components. Within Table 2 the investigated equivalence ratio Φ , fuel composition, temperature and the laminar burning velocity S_L for the experimental data is given.

TABLE 2 LAMINAR BURNING VELOCITY S_L for DIFFERENT FUEL COMPOSITION and EQUIVALENCE RATIOS Φ

case	fuel composition [vol. %]		equivalence ratio Φ	laminar burning velocity [cm/s]	Temperature T_{gas} [K]
	CH ₄	H ₂			
1	100	-	0.7 - 1.3	16.51 - 36.24	298
2	95	5	0.8 - 1.3	24.29 - 37.95	298
3	90	10	0.8 - 1.3	25.05 - 39.22	298
4	H ₂	N ₂			
5	30	70	1.0 - 1.5	46.97 - 58.11	298
6	25	75	1.0 - 1.5	29.55 - 37.51	298
7	20	80	1.0 - 1.5	12.76 - 19.54	298
7	18	82	1.0 - 1.5	8.55 - 13.51	298

A comparison between the laminar burning velocities of methane-air mixtures at ambient initial conditions from the current measurements and from the literature based on various experimental techniques is shown in Fig. 3. The current results of the laminar burning velocities experiments are close to the literature. They were reported by using stationary flames and heat flux measurements or counter-flow flames. Also, a numerical approach with USC II for methane-air is shown, therefore it could be seen that the experimental data in the $\Phi > 1$ area mostly have lower burning velocities than the numerical results. In the area $\Phi < 1$ the numerical approach fit the experimental data better. The trend of the curve and the maximum point could be simulated very well.

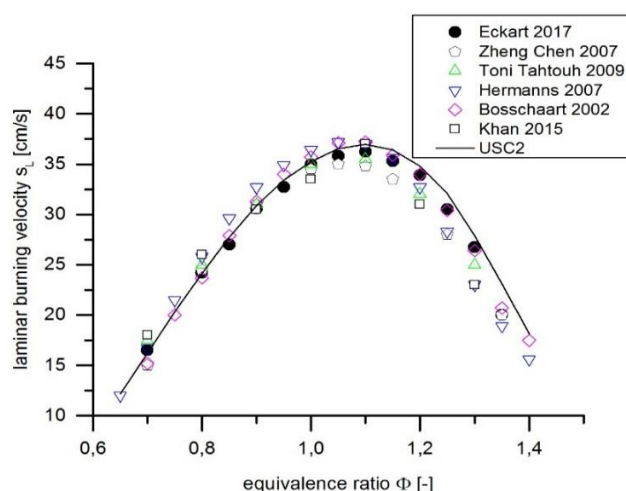


Fig. 3 Validation of present experimental data versus published experimental data and kinetic simulations for CH₄/air burning velocity [8] [20] [21] [22] [23]

After verification of the method with literature given values, the influence of inert gases was examined. Therefore, methane was diluted with different percentages of carbon dioxide and nitrogen.

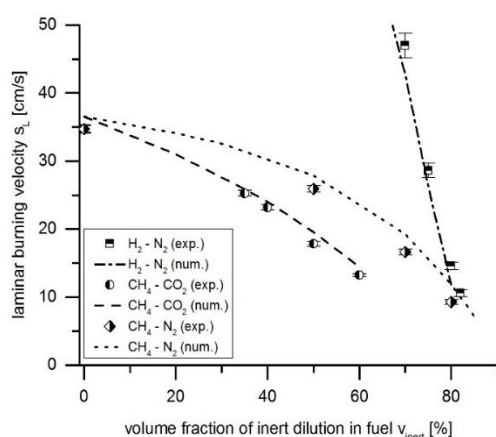


Fig. 5 Effect of dilution (CO_2 and N_2) on the laminar burning velocity, experiments and numerical investigation at $\Phi = 1.0$

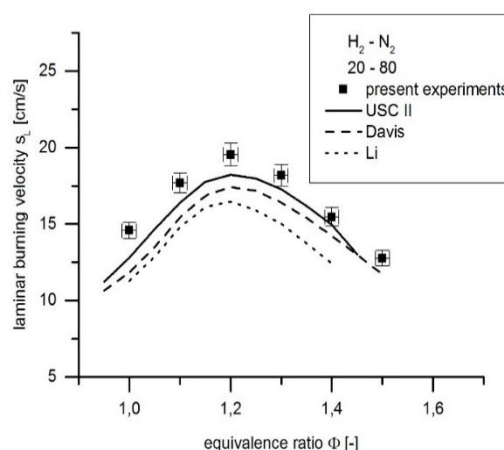


Fig. 4 Comparison of different reaction mechanism with experimental data for $\text{H}_2\text{-N}_2$ mixture

Fig. 5 shows the effect of dilution for the laminar burning velocity for $\text{CH}_4\text{-N}_2$, $\text{CH}_4\text{-CO}_2$ and $\text{H}_2\text{-N}_2$ gas mixtures. Assuming that a fuel gas content of zero volume percent fuel has a laminar burning velocity of zero, it seems possible to construct a nearly linear curve for the $\text{CH}_4\text{-CO}_2$ gas mixtures. For the measured $\text{H}_2\text{-N}_2$ gas mixtures the results also show a nearly linear curve but no zero-crossing. Only for the burning velocities of $\text{CH}_4\text{-N}_2$ gas mixtures the function shows no linear tendency. This result suggests that there is an interaction of the gas components in the $\text{CH}_4\text{-N}_2\text{-O}_2$ mixture that affects the physical effects, which is also addressed by work from Kondo et al. [24]. It could be shown, that the numerical results cannot predict the experimental results exactly. The numerical results (GRI 3.0) slightly overestimates the burning velocities in case of $\text{CH}_4\text{-N}_2$ and $\text{CH}_4\text{-CO}_2$ mixtures.

For the $\text{H}_2\text{-N}_2$ mixtures three reaction mechanisms were compared, as given in Fig. 4. The USC II underestimates the experimental results slightly, the Li and Davis mechanisms have a much bigger deviation from the present experimental data and will not be used in the further numerical simulations. In all three cases it is possible to cover the trend of the laminar burning velocity.

B. Effect of dilution of hydrogen in nitrogen and methane

After showing the effect of rising fuel content in the mixtures and influence of inert components, the main focus will be on hydrogen mixtures. For the German and European gas grid the influence of hydrogen in methane is one of the most important topics for the future. In our experiments nitrogen diluted hydrogen mixtures were investigated. In respect to the upper limits of the burner setup, the maximum burning velocity has to be below 65 cm/s. Therefore, the maximum share of hydrogen/air cannot exceed 30 vol. %. If higher shares want to be tested the amount of oxygen in the mixture can be reduced, which was already done in some experiments from De Goey and Maaren [25]. In the present experiments the hydrogen share between 18 to 30 vol. % were analysed.

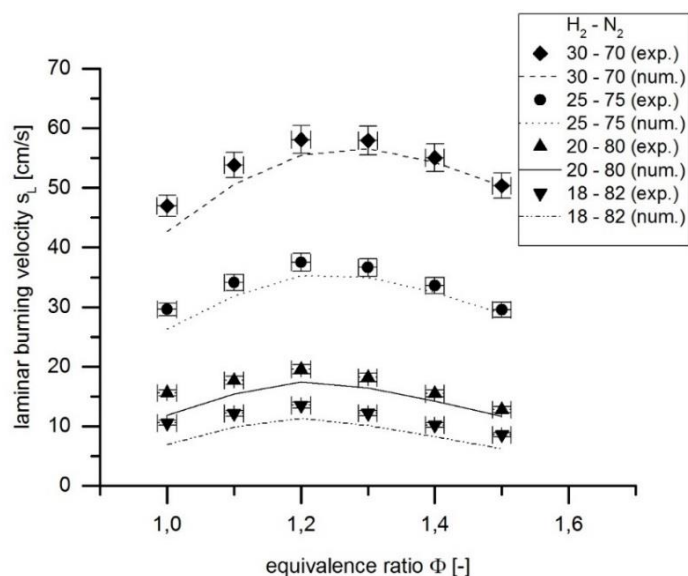


Fig. 6 Validation of present experimental data versus simulations (USC II) for H₂-N₂/air burning velocity

Fig. 6 depicts that for H₂-N₂ mixtures the laminar burning velocities increases with the share of fuel. For combustion of pure H₂, the maximum burning velocity is around 385 cm/s [26]. The maximum laminar burning velocity of the 18%-H₂/82%-N₂ at 298K increases from around 11 cm/s to 30%-H₂/70%-N₂ to a maximum velocity of 59 cm/s. Results from similar gas mixtures investigated by Slepterevalso et al. [27] shows a similar behaviour. The maximum in all curves is always located in the area of $\Phi = 1.2$, which is clearly higher as the equivalence ratio of the maximum point for pure methane. With rising delusion of hydrogen this maximum seems to shift to higher equivalence ratios, which was also examined by Hermanns [25]. Pure hydrogen has the maximum laminar burning velocity at an equivalence ratio of 1.7. As seen in Fig. 5 the trend of the burning velocity in the region between 1.0 and 1.5 is well reproduced by numerical simulations using the USC-II mechanism. In the area of $\Phi = 1$ the simulations slightly underestimate the experimental results of the heat flux burner, whereas it nearly matches for higher equivalence ratios. The difference between the numerical approach and the experimental results is between 0.5% and 7.1 %. Also it could be shown that an additional 5 vol. % of H₂ to 25vol% has more impact, than reduce it 5 vol. % to 20% H₂. With a rising of 5vol% the burning velocity at $\Phi = 1$ rises from 29.6 cm/s to 46.9 cm/s (at 30%H₂-70%N₂). At the dilution with only 20vol% hydrogen the maximum burning velocity drops to 15.57 cm/s.

Further experiments deal with the current possible share of hydrogen in the gas grid. Therefore, the burning velocity for a methane hydrogen mixture was analysed.

TABLE 1 COMPARISON of EXPERIMENTAL DATA for METHANE/HYDROGEN LAMINAR BURNING VELOCITY, the MEASUREMENTS WERE DONE at 298K with DIFFERENT EQUIVALENCE RATIOS and with 20.9% OXYGEN in OXIDIZER. ALL POINTS are GIVEN in cm/s.

equivalence ratio	CH ₄ 100-0 H ₂ [vol.%]	CH ₄ 95-5 H ₂ [vol.%]	CH ₄ 90-10 H ₂ [vol.%]
0.7	16.51	-	-
0.8	24.23	24.29	25.05
0.9	30.58	31.03	32.63
1.0	34.97	36.31	37.91
1.1	36.24	37.95	39.22
1.2	33.89	34.91	36.40
1.3	26.75	28.01	29.57

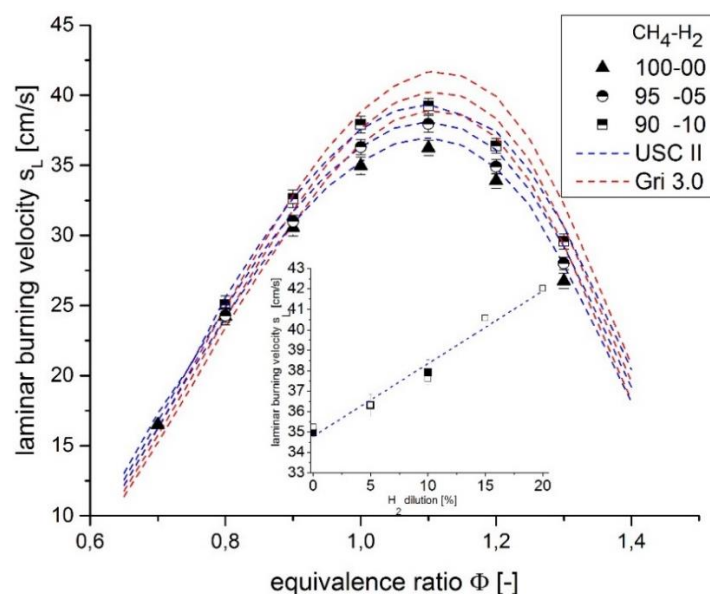


Fig. 7 Calculated laminar burning velocity from two mechanisms versus present experiments at 298K, the equivalence ratio for various percentage of hydrogen, in the small diagram the numerical (open symbols) and experimental (closed symbols) laminar burning velocity for the equivalence ratio of 1,0 with different hydrogen dilution

The effects of hydrogen addition on the laminar burning velocity at temperature 298 K and atmospheric pressure are shown in Fig. 7. The Hydrogen content in the fuel is varied from 0% to 10% in volume for the experiments and from 0% to 20% in the simulation. In the case of hydrogen, the measurements have to be performed very carefully, because of the flame flashes back for the stoichiometric and rich conditions. To prevent the possibility of flashback, it is necessary either to increase the inlet velocity of the gas flow, to reduce the quenching diameter of the holes in the burner plate or to decrease the area of the exit burner. In the experiments it could be shown that with a rising amount of hydrogen in the mixture, the burning velocity also rises at all tested equivalence ratios. This is due to the high mass and thermal diffusivity of hydrogen in air and its high reactivity compared to methane. This increase of laminar burning velocity would lead to very different performances, like temperature shift and flame length, in real combustion systems. In the area $\Phi < 1$ the experimental results are closer to each other, then in the area $\Phi > 1$. It seems that the hydrogen has a stronger influence in the burning velocity $\Phi > 1.1$. The maximum laminar burning velocity could be discovered in the region around $\Phi = 1.1$. Because of the tested interval of $\Phi = 0.1$ a closer diagnostic is not possible at the moment in will be determined experimentally in following articles. To show a possible trend of shifting behavior with the maximum point of the laminar burning velocity.

The burning velocity of pure methane at 298K and $\Phi = 1.1$ rises about 4.7 % when only 5 % of hydrogen are added. With a hydrogen amount of 10% this rises by 8.2% of the original value. The experimental results, shown in Table 3, were also compared with 2 reactions mechanism. Both of them, GRI 3.0 and USC II, can predict the trend of the curve really good. In the area of $\Phi > 1.3$ and $\Phi < 0.8$ both mechanics show nearly the same results for the burning velocity. Therefore, the USC II mechanisms overestimates the experimental data. In the range from an equivalence ratio of 0.9 to 1.1 the numerical results predict the experimental data and the general trend quite well. Also, the position of the maximum value is represented in the same manner. The mechanism GRI 3.0 overestimates the present experimental data from an equivalence ratio of 0.8 to 1.3 clearly. It also can be shown that the numerical results show a quasi linear trend and the experimental result follow this clearly in the area from 0% to 10% hydrogen dilution.

V. CONCLUSIONS

In the present investigations on hydrogen gas mixtures the laminar burning velocity was investigated as one of the most important combustion properties to numerical reaction mechanisms. These gas mixtures have been tested under atmospheric conditions in a heat-flux burner setup. The burning velocity is very important for thermos-process technologies in the future in order to make combustion processes even more efficient and save. These gases consist primarily of combustible components such as hydrocarbons and hydrogen.

The heat-flux burner was adapted with a conditioning and preheating system for gaseous fuels. The experimental system was successfully validated with the pure reference fuel methane by comparing to previously published literature data for the laminar burning velocity. For all measurements the experimental data were compared to numerical predictions.

For comparison of experimental and simulated data the standard deviation (always in terms of cm/s) was used as a measure for the variance to the average value of the laminar burning velocity. It could be shown that the laminar burning velocity of H₂/N₂-air flames at 298 K and atmospheric pressure varying the H₂/N₂ ratio. The ratios range from 18%-H₂/82%-N₂ with a maximum laminar burning velocity of $S_{L, \text{Max}} = 13.5$ cm/s for up to 30%-H₂/70%-N₂ with $S_{L, \text{Max}} = 58.1$ cm/s. It could be shown that under 17%-H₂ the flame becomes unstable and not measurable with the used system. On the other hand, the laminar burning velocity increases for higher H₂/N₂ ratios. Also, the influence of hydrogen in methane was tested. Therefore, the results clearly show that with a rising H₂ concentration in the mixture not only the laminar burning velocity rises but also the point of the maximum velocity shifts to a higher equivalence ratio. Within all experiments for the laminar burning velocity only a small experimental uncertainty can be seen. Furthermore, the experimental results show a good agreement with the published experimental data and also with the numerical predictions for these fuel mixtures. The numerical results from USC II mostly slightly underestimate the experimental data for hydrogen/nitrogen mixtures whereas an overestimation was seen for methane/hydrogen mixtures. Furthermore, all the tested reaction mechanisms predict the correct trend of the laminar burning velocity when varying the hydrogen fraction. Nevertheless, there is need for an improved description of the chemical kinetics for these mixtures in lower share of combustion gases and lower equivalence ratios.

In the future the experiments of the presented matrix for hydrogen containing fuel blends will be expanded. More mixtures with a hydrogen share from 15 to 40 vol. % will be investigated to allow a better understanding for these fuels. The results will be also compared with numerical simulations.

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