

EFFECT OF TEMPERATURE ON LAMINAR FLAME VELOCITY FOR HYDROGEN-AIR MIXTURES AT REDUCED PRESSURES

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ABSTRACT

The work was done with respect to hydrogen safety of ITER vacuum vessel in cases of loss of cooling and loss of vacuum accidents. Experiments were conducted at sub-atmospheric pressures from 1 bar to 200 mbar and elevated temperatures up to 300 °C. Hydrogen concentration was changed from lower to upper flammability limits in all the range of pressures and temperatures. The experiments were performed in a spherical explosion bomb equipped with two quartz windows. The flame propagation velocity was measured using pressure method and high speed shadow cinematography. The theoretical flame velocities were calculated by Cantera code using Lutz and Mueller mechanisms. The influence of the initial temperature and pressure conditions on the laminar flame speed S_L , overall reaction order n and Markstein length L_M are presented in this work and compared with the results of a theoretical model.

1.0 INTRODUCTION

Under hypothetical Loss-of-Cooling-Accident (LOCA) scenarios of International Thermonuclear Experimental Reactor (ITER), hydrogen may be produced due to the reaction of hot beryllium panels with cooling water. Another source of hydrogen in ITER sub-modules is hydrogen released from cryo-pumps. Air ingress from ambient atmosphere into the Vacuum Vessel (VV) may lead to formation of combustible hydrogen-air mixture. Ignition of the mixture by internal ignition source and its combustion in the vacuum vessel may generate high pressure and thermal loads. The hydrogen hazard in LOCA sequences was analysed systematically for vacuum vessel, cryo-pumps and neutral beam systems [1, 2].

Typical ITER relevant initial conditions may vary in the range of a few Pa to atmospheric pressure (1 bar) and temperature from cryogenic to 400 °C. Depending on hydrogen inventory and air ingress the initial pressure may be changed from 50 mbar to atmospheric pressure. The flammability limits of hydrogen-air mixtures at ambient temperature of 293K and reduced pressure up to 30 mbar are shown in Fig. 1 for two different ignition sources [3].

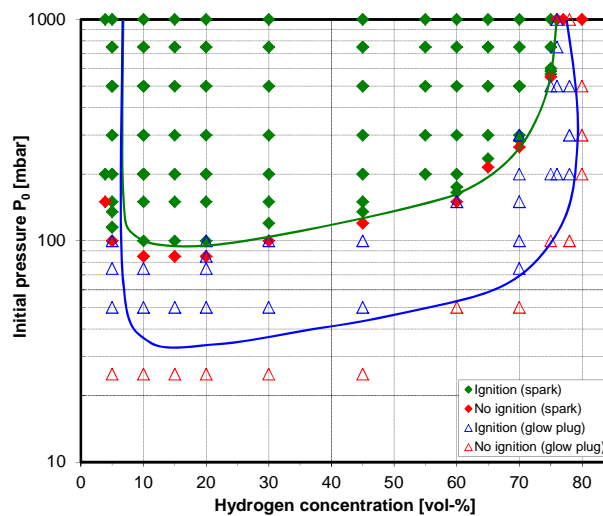


Figure 1. Flammability limits of hydrogen–air mixtures at sub-atmospheric pressures ($T = 293$ K) [3].

The picture shows that the Lower Flammability Limit (LFL) is not sensitive to the initial pressure. It remains at about 4% hydrogen in air, as it is at atmospheric pressure, independent of the initial pressure. The Upper Flammability Limit (UFL) was found to be quite sensitive to the initial pressure in the range of 50–1000 mbar. The UFL extends from 75% H_2 at 1000 mbar to 80% H_2 at pressures in the range of 200–500 mbar, and then below 200 mbar the UFL reduces from 80% H_2 to 50% H_2 at a pressure of 50 mbar. The lower flammable pressure of hydrogen-air mixture is about 30–40 mbar.

Another problem to ignite the hydrogen–air mixtures at reduced pressures is the dependency of Minimum Ignition Energy (MIE) on initial pressure p_0 and temperature T_0 as following [4]:

$$E_{ig,min} \sim T_0^{-2} \cdot p_0^{-2}, \quad (1)$$

This means that it will need more energy to ignite the same mixture at reduced pressures. Figures 2 demonstrate the minimum ignition energy for hydrogen-air mixtures as a function of initial pressure and stoichiometric ratio of the mixture. We may roughly evaluate from both pictures that to ignite a hydrogen-air mixture near the flammability limits it needs almost 1 J of ignition energy.

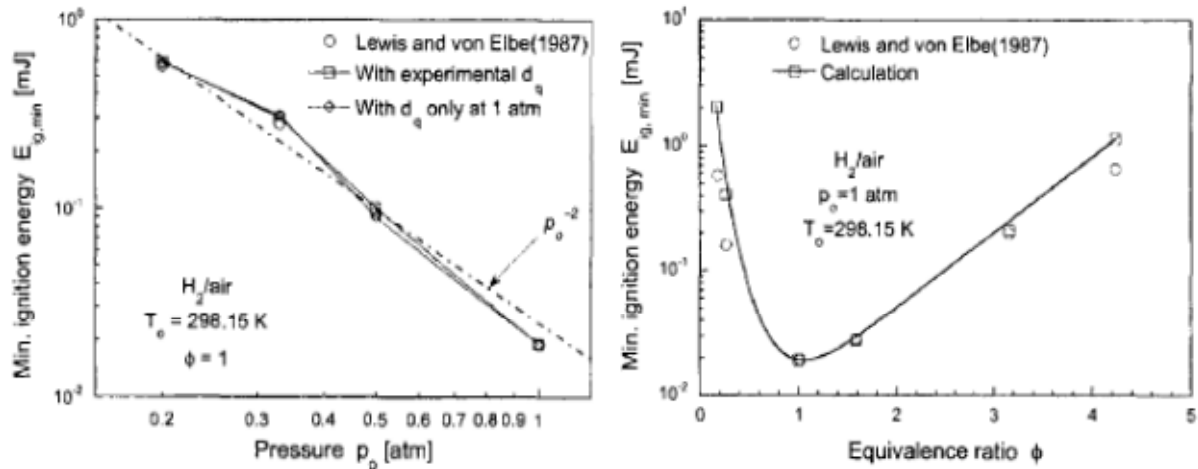


Figure 2. Minimum ignition energies (MIE) of stoichiometric hydrogen/air mixture for $T_0=298$ K as a function of pressure (left) and hydrogen concentration at $p_0 = 1$ atm (right) [5].

Effect of initial temperature on flammability limits of hydrogen-air mixtures is shown in Fig. 3. The picture accumulates published data [6-7] on flammability limits at ambient pressure in wide range of temperatures even up to the auto-ignition temperature [8]. The auto-ignition limit was additionally calculated in [9] using Maas-Warnatz mechanism and INSFLA code. The dependency of lower and upper flammability limits on temperature is almost linear. It extends from 4–75 % H_2 at 20°C to 1.5–87.6 % H_2 at 400°C [10].

The problem of air ingress into the ITER vacuum vessel filled with hydrogen at reduced pressure was investigated in [11]. As an example for 20 kg of hydrogen inventory, during an air ingress the internal pressure changes from 200 mbar to atmospheric pressure. Hydrogen concentration will simultaneously change from 100 % H_2 in the beginning to 20 % H_2 at the end. Below the upper flammability limit UFL=80 % H_2 , the gaseous mixture becomes flammable.

Within the flammability limits for hydrogen-air mixtures flame may propagate in different regimes from quasi-laminar to sonic deflagration and even detonations. Critical conditions for flame propagation regimes in confined volumes depending on geometry, scale and initial mixture properties are written in papers [12-13]. As mentioned in these papers, the main mixture properties governing the flame propagation regime from subsonic quasi-laminar to supersonic flame and then detonations are expansion ratio, laminar burning velocity, speed of sound in reactants and products, detonation cell size and many others. Expansion ratio gives a thermodynamic potential for effective flame acceleration to sonic speed, the laminar burning velocity and speed of sound characterize dynamic of the flame propagation. Hazard potential of combustible mixture in terms of pressure and thermal loads will strongly depend on flame propagation velocity [12, 14].

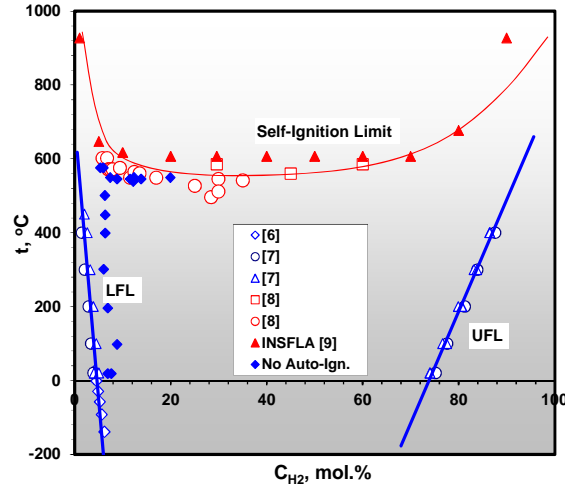


Figure 3. Flammability limits of hydrogen –air mixtures at ambient pressure ($p_0 = 1$ bar) and different temperatures up to the auto-ignition limit.

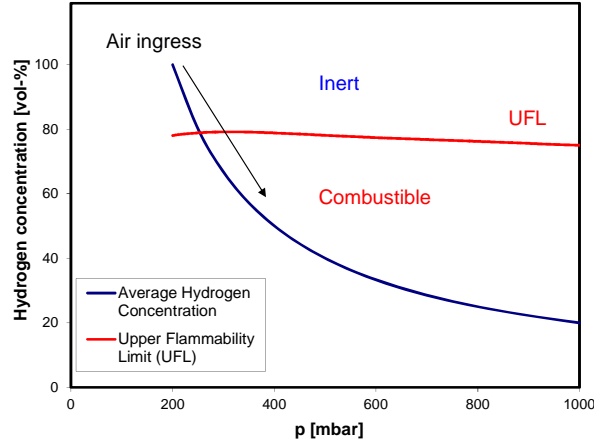


Figure 4. State of hydrogen-air composition inside the ITER vacuum vessel for 20 kg of hydrogen inventory during an air ingress until internal pressure will be equal to atmospheric (1 bar).

The laminar burning velocity (LBV) is one of important overall characteristics of the reactivity of combustible mixtures. It is usually used as a measure of the combustion rate in numerical models to predict dynamics of the combustion. The LBV can be used to evaluate the laminar flame thickness δ , the overall activation energy E_a , and the reaction order n , for the dimensionless analysis of turbulent combustion and to estimate flame stability and flame acceleration potential.

According to the Zeldovich - Frank-Kamenetskii theory [15], the overall activation energy E_a represents the sensitivity of the laminar burning rate to the flame temperature as follows

$$S_L(T_b) \propto S_L \cdot \exp\left(-\frac{E_a}{2 \cdot T_b}\right), \quad (2)$$

where $S_L(T_b)$ and S_L are laminar burning velocities at a given flame temperature T_b and at ambient temperature T_0 . The pressure dependence of the laminar velocity as a function of the reaction order n can be written in simplified form:

$$S_L(P) \propto S_L(P_0) \cdot \left(\frac{P}{P_0} \right)^{\frac{n}{2}-2}, \quad (3)$$

The overall reaction order n indicates the pressure effect on the combustion rate. For instance, if reaction order $n < 2$, the laminar burning velocity will decrease with an initial pressure increase.

This is why the laminar velocity of hydrogen-air combustion at reduced pressures and different temperatures is an important issue from a practical point of view. The problem related to the hydrogen safety of ITER sub-modules is that the only several experimental works are recently known with the data on laminar burning velocity of hydrogen-air mixtures at reduced pressures and normal temperature [3, 16-21]. Figures 5-7 show a comparison of laminar burning velocity of hydrogen-air mixtures at sub-atmospheric pressures in these papers. One can see that laminar flame speed increases with pressure increase for hydrogen-air mixtures within the range of 20 – 60% H_2 . Out of this range, for very lean and very rich hydrogen-air mixtures, the pressure has a suppressing effect on laminar flame velocity. Such a behavior was shown to depend on the overall reaction order [3, 21] according to Eq. (3). It was experimentally found, the overall reaction order $n > 2$ within the range 25-65% H_2 leads to promoting effect of pressure on laminar flame speed [3]. Almost the same range of hydrogen concentrations with reaction order $n > 2$ was calculated in [21] using modified Yetter mechanism [22]. As shown in [20], the mechanism of Mueller et al. [23] predicted reasonably well experimental results of the laminar burning velocities at lean and rich equivalence ratios; however it tended to overestimate experimental values of S_L at ultra-rich equivalence ratios.

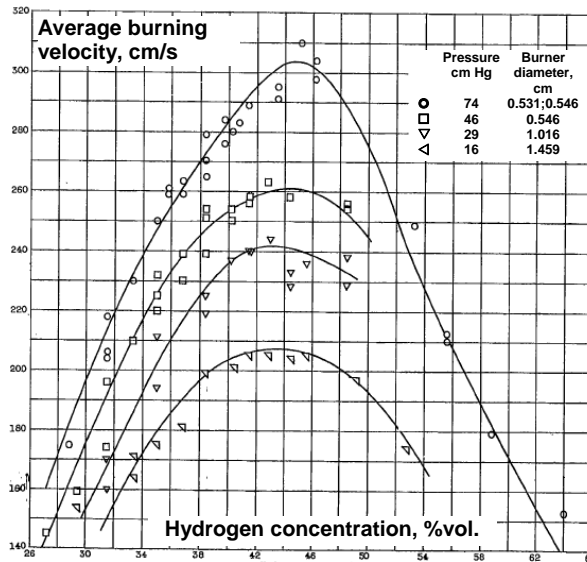


Figure 5. Burning velocities of hydrogen-air flames [16]: (1) 986 mbar; (2) 613 mbar; (3) 387 mbar; (4) 213 mbar.

Another effect of the pressure is related to the flame stability, specifically to the influence of flame front stretch on laminar flame speed. According to Eqs. (4) –(5) assuming a linear correlation, this effect can be specified by Markstein length L_M :

$$S_L = S_{L,s} - L_M \cdot K, \quad (4)$$

where S_L and $S_{L,s}$ are the stretched and free of stretch laminar flame velocities, A is the flame area, K is the stretch rate calculated for spherical case as follows

$$K = \frac{1}{A} \frac{dA}{dt} = \frac{2}{r_b} \frac{dr_b}{dt}, \quad (5)$$

It was found numerically in [21] and experimentally in [3] that for all the concentrations, the Markstein length increases with decreasing initial pressure. Lean mixtures with less than 25% H_2 ($\phi < 0.8$) have negative Markstein lengths at sub-atmospheric pressures, while for stoichiometric and rich mixtures positive Markstein lengths were measured. That is that for lean mixtures the stretch effects cause an acceleration of the curved flame front to the planar, while for stoichiometric and rich mixtures they cause a deceleration of the flames approaching to planar.

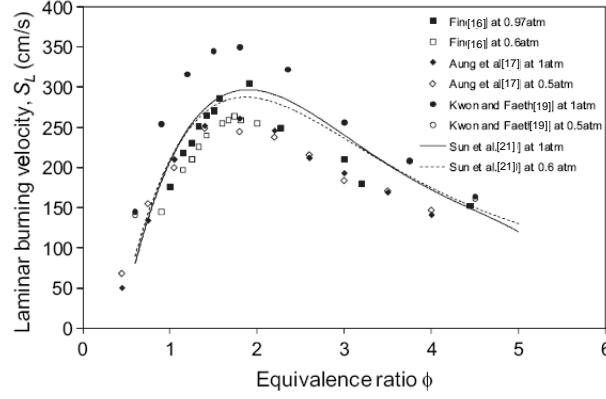


Figure 6. Laminar burning velocities of H_2 /air flames at atmospheric and low pressures. Markers: experimental results [16], [17] and [19]; lines: numerical results [21].

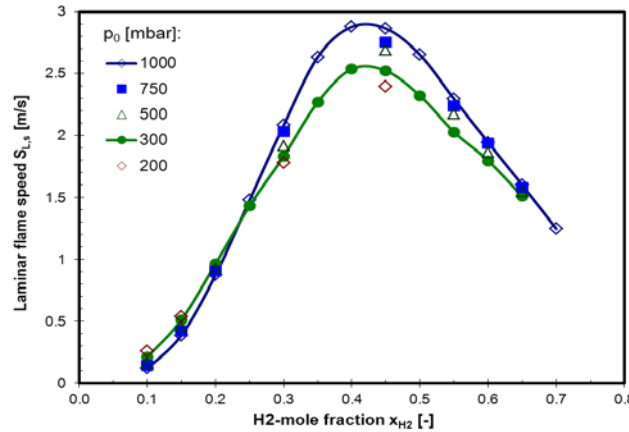


Figure 7. Unstretched laminar flame speeds vs. hydrogen concentration at different pressures and initial temperature of 290K [3].

There is a lack of similar experimental data for sub-atmospheric pressures at elevated temperatures above the normal temperature. Especially it is very interesting how the laminar flame behaves under initial conditions in the vicinity of self-ignition limit when initial temperature is of the order 300-500 °C relevant to ITER operating conditions.

Objectives

The objective of the current work is to obtain experimental data on the laminar burning velocities in hydrogen-air mixtures at reduced pressures in wide range of temperatures from 20 to 300 °C. With these data the possibility of combustion events and their regimes can be evaluated for hydrogen safety related to ITER applications using CFD simulations.

2.0 CALCULATIONS

Numerical calculations of the laminar burning velocities were obtained using Cantera code [24] for

unstretched (plane) flames. The main problem of the calculations was to achieve convergence of the numerical solution for certain initial conditions close to flammability limits. To calculate laminar burning velocities of hydrogen-air mixtures as function of initial pressures and temperatures, two different H/O reaction mechanisms based on the Lutz scheme [25] and Mueller mechanism [23], were used in this work.

As an example, the effect of initial pressure on the laminar burning velocity of H₂-air mixtures at different temperatures for Mueller mechanism [23] is shown in Fig. 8. It was found that for very lean (< 20% H₂) and very rich (> 65-80% H₂) mixtures the initial pressure has a suppressing effect on the laminar burning velocity. In contrary, for the middle hydrogen concentrations (20 – 65 % H₂), the laminar flame velocity increases with a pressure increase from 0.1 to 1 bar. As usually such behaviour of laminar flame velocity against initial pressure associates with an overall reaction order. According to Eq. (3), this means, the overall reaction order n is higher than 2 in the range of middle hydrogen concentrations.

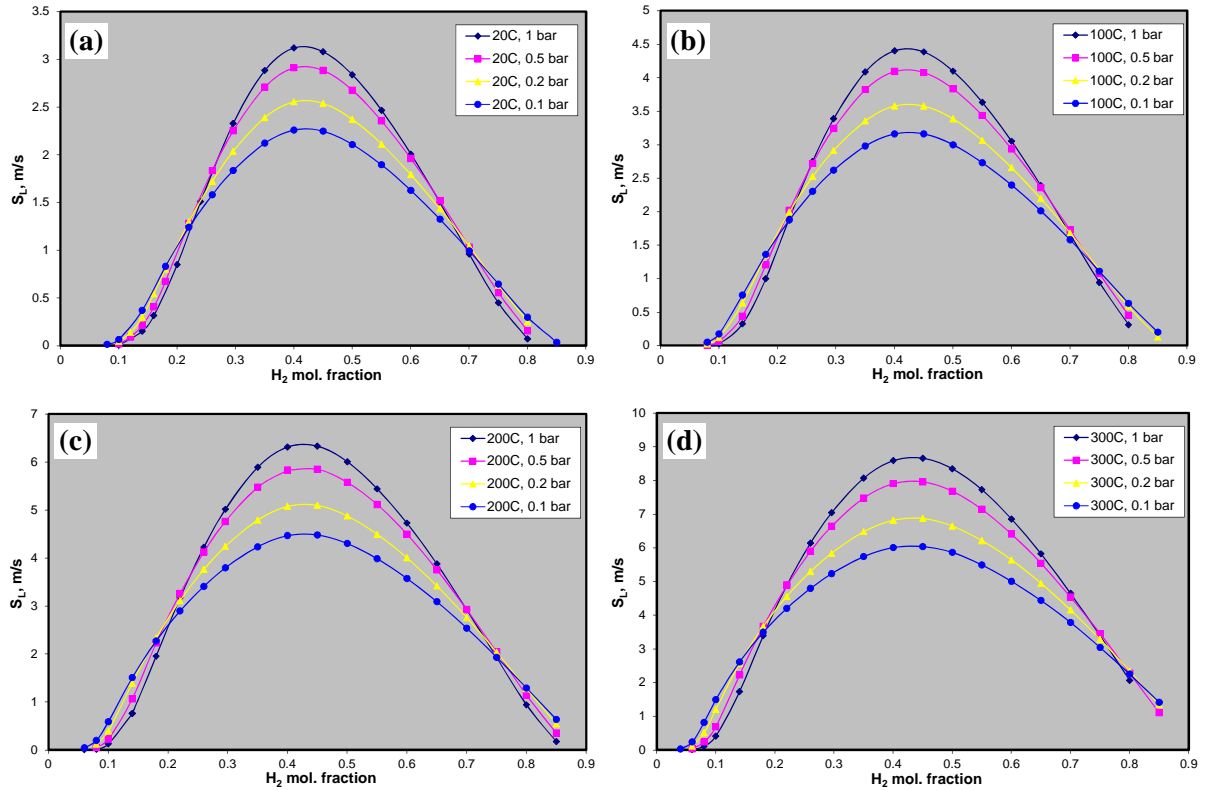


Figure 8. Calculated laminar flame velocities vs. hydrogen concentration at different pressures and temperatures (Mueller mechanism): (a) - 20°C; (b) - 100°C; (c) - 200°C; (d) - 300°C.

Figure 9 shows a dependence of overall reaction order by Eq. (6) vs. H₂ concentration at two temperatures.

$$n = 2 \frac{\partial \ln(S_L)}{\partial \ln(p)} + 2 \quad (6)$$

Similar behavior demonstrate calculated dependencies of overall reaction order against hydrogen concentration for other initial temperatures of 100 and 200 °C. Almost the same curve was experimentally obtained in paper [3] and numerically calculated in paper [21] for 20 °C. It was found in the current work, similar to paper [21], that the overall reaction order increases at lower pressures in the vicinity of upper and lower flammability limits.

Data of numerical calculations using Lutz and Mueller mechanisms for sub-atmospheric pressure of 200 mbar and elevated temperature up to 300 °C are summarized in Fig. 9. Both mechanisms

give quite similar data on laminar flame velocity at reduced pressure of 200 mbar with a difference that Lutz data a bit higher, especially at higher temperatures. Taking into account the opinion in paper [20] that the Mueller mechanism is one of the best suitable for sub-atmospheric pressures, we have chosen Mueller mechanism as a basic for further data analysis.

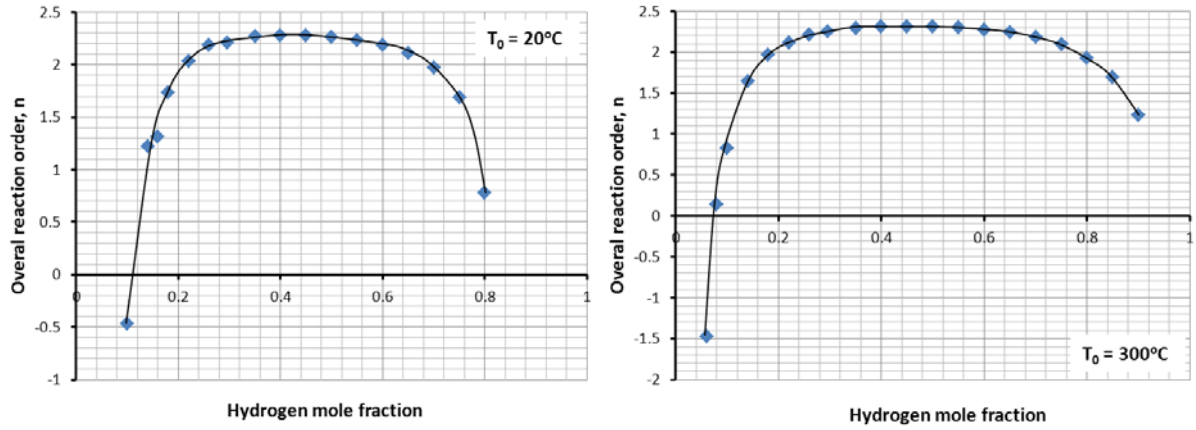


Figure 9. Calculated overall reaction order for $T_0=20^\circ\text{C}$ and $T_0=300^\circ\text{C}$ (Mueller mech.)

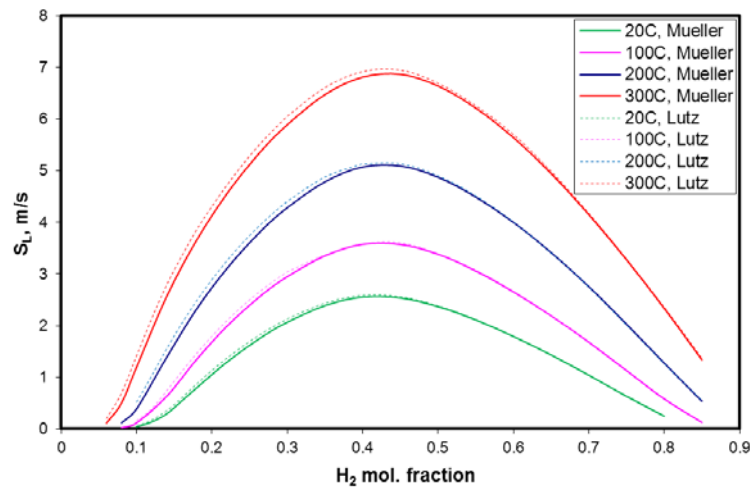


Figure 10. A comparison of calculated unstretched laminar flame speeds vs. hydrogen concentration at different temperatures and initial pressure of 200 mbar: Mueller mech. (solid); Lutz scheme (dash line)

3.0 EXPERIMENTAL DETAILS

Experiments on laminar burning velocity were conducted in a spherical stainless steel explosion bomb having an inner diameter of 25 cm ($V = 8.2 \text{ dm}^3$). The explosion bomb with a wall thickness more than 34 mm and two quartz windows for optical observations, was designed to withstand a static pressure up to 800 bar and a temperature up to 300°C (Fig. 11).

Premixed hydrogen – air mixtures within the flammability limits 5-75 % H_2 were used in the tests. The mixtures to be tested were prepared by mass flow rate controller. Each component of a gas mixture was heated up to the required temperature separately before the mixing inside the test volume. The mixture quality was controlled by a gas analyzer via bypass line before filling of the test volume.

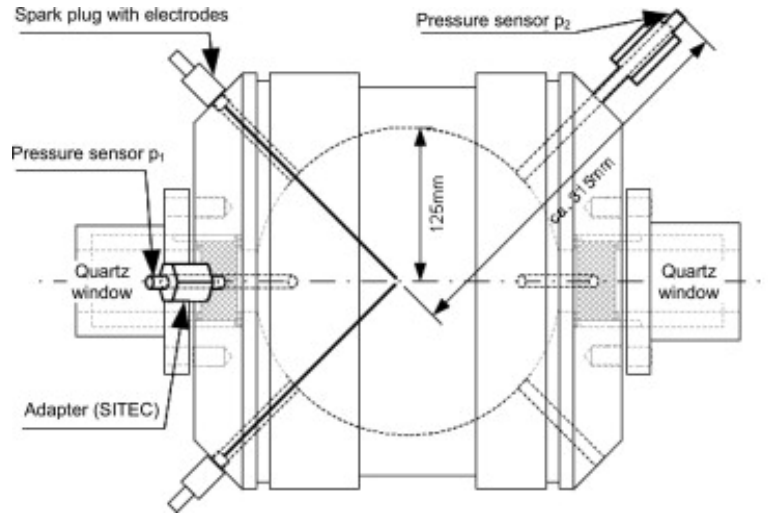


Figure 11. Explosion chamber for laminar velocity evaluation.

Combustion experiments have been done at initial temperatures of 20, 100, 200 and 300 °C and pressures of 200, 500 and 1000 mbar. An insulated explosion bomb was heated by 4 kW heating elements (Horst HT30) with a control by thermocouples. The gas temperature was measured using a NiCr-Ni thermocouple of 0.2 mm thickness installed at 6 cm from the wall inside the test mixture. The temperature control system has the initial temperature deviations ± 0.5 °C. The spatial non-uniformity of the temperature was less than ± 1 C.

A calibrated spark plug or glow plug were used to ignite the test mixture. For some mixtures close to the flammability limit, the minimum ignition energy could be about 0.5 – 1.0 J [4]. Experiments showed that both ignitors give reliable data on the flame velocity. Figure 12 shows experimental data of our preliminary experiments on minimum ignition energy as function of initial pressure and hydrogen concentration. The data are in perfect agreement with paper [4].

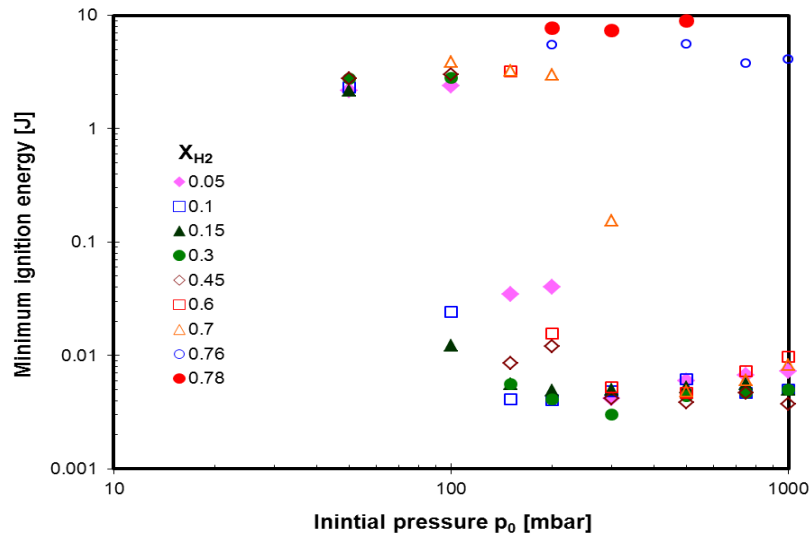


Figure 12. Minimum ignition energy as function of initial pressure and H₂ concentration ($T_0 = 20$ °C).

Measurements of laminar burning velocities were made using a high speed shadow cinematography (up to 20 000 fps) and pressure transducers. Two piezo-electric pressure transducers with air cooling system operating with a temperature limit of 350 °C and with a measuring pressure range of 0 – 3.45 – 34.5 bar (PCB 112A22 and 113A36) were used to measure the combustion pressure.

4.0 RESULTS AND DISCUSSION

Free of stretch laminar flame velocity was calculated using Eq. (4)-(5). A high speed schlieren technique and spherical bomb pressure method were used to measure flame radius vs. time. For the pressure method assuming spherical shape of the flame, the flame radius r_b and stretched flame speed S_L were evaluated in current work by Dahoe [26]:

$$\frac{r_b(t)}{R} = \left[1 - \left(\frac{p_0}{p} \right)^{1/\gamma} \frac{p_e - p}{p_e - p_0} \right]^{1/3} \quad \text{and} \quad (7)$$

$$S_L = \frac{R}{3} \frac{1}{p_e - p_0} \left(\frac{p_0}{p} \right)^{1/\gamma} \left[1 - \left(\frac{p_0}{p} \right)^{1/\gamma} \frac{p_e - p}{p_e - p_0} \right]^{-2/3} \frac{dp}{dt} \quad (8)$$

Figure 13 shows a comparison of free of stretch laminar flame velocity S_L with numerical calculations by Cantera code and Mueller mechanism [23] for different pressures and temperatures in wide range of concentrations. One can see very good consistency of optical and pressure measurements in current work. Our calculations give much bigger discrepancy with experimental data. In contrary to work [20], the Mueller mechanism [23] is quite close or underestimated the experimental values of S_L at lean and rich hydrogen concentrations. In the range of middle hydrogen concentrations the calculations overestimated the experimental values of S_L . We also may see that the lower the initial temperature the better is agreement of calculations to experimental data.

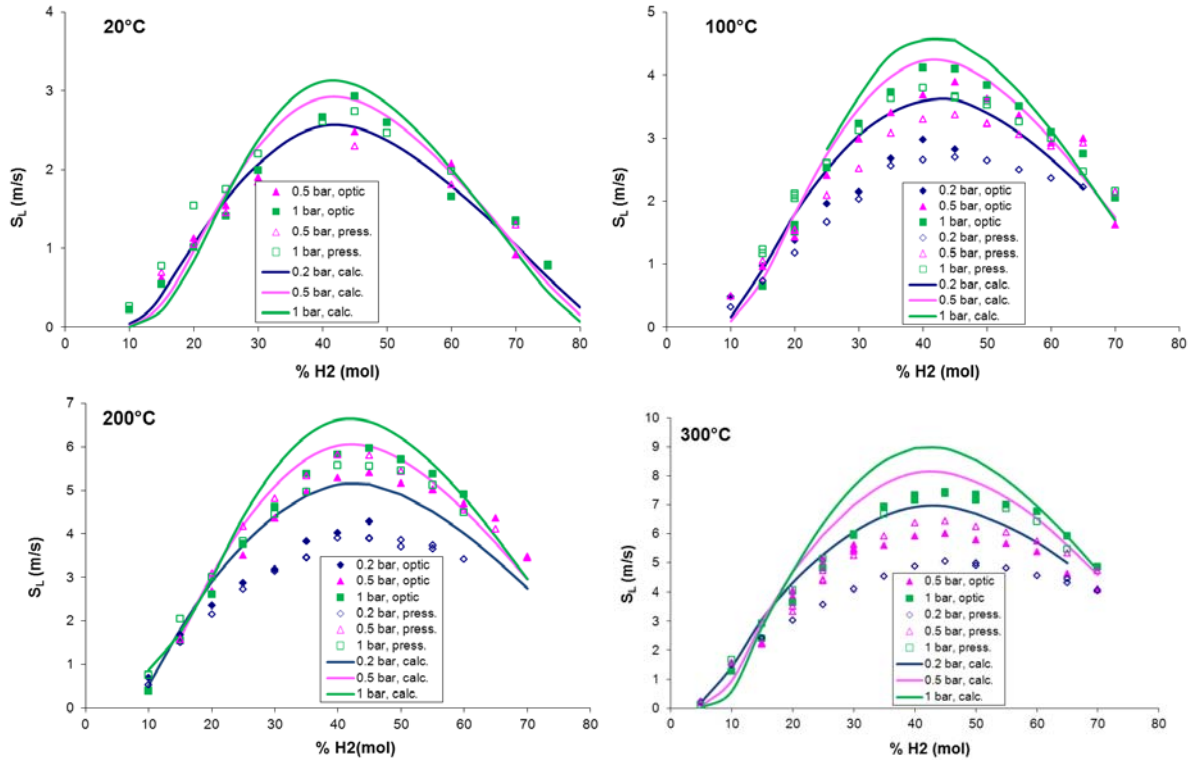


Figure 13. Comparison of experimental and calculated laminar flame velocities at different temperatures and sub-atmospheric pressures

Additionally to the laminar flame speed the Markstein length L_M was extracted from experimental data processing. Figure 14 (left) shows a general behavior of Markstein length at different hydrogen concentrations as function of initial pressure at 200 °C. Similar behavior was found for other

temperatures. The Markstein length characterizes a flame sensitivity to diffusional-thermal instability. A positive value indicates the flame is stable to the diffusional-thermal effect, whereas a negative Markstein length indicates the flame surface will be distorted due to diffusional-thermal instability leading to the acceleration of flame speed and the formation of a cellular structure. As we may see in Fig. 13 (left), the threshold positive-negative L_M shifts from very lean to higher hydrogen concentrations with reduced pressures. A positive L_M at lower pressure, for instance, leads to changing of very fine cellular flame structure of 15 %H₂-air mixture at 1 bar ($L_M < 0$) to almost smooth flame surface at 200 mbar ($L_M > 0$) (Fig. 14 a,b). Effect of elevated temperature is not so significant but, nevertheless, according to the temperature dependence of Markstein number L_M (Fig. 13, right) the cellular structure at 300°C becomes larger compared to very fine structure at 20°C (Fig. 14 b,c).

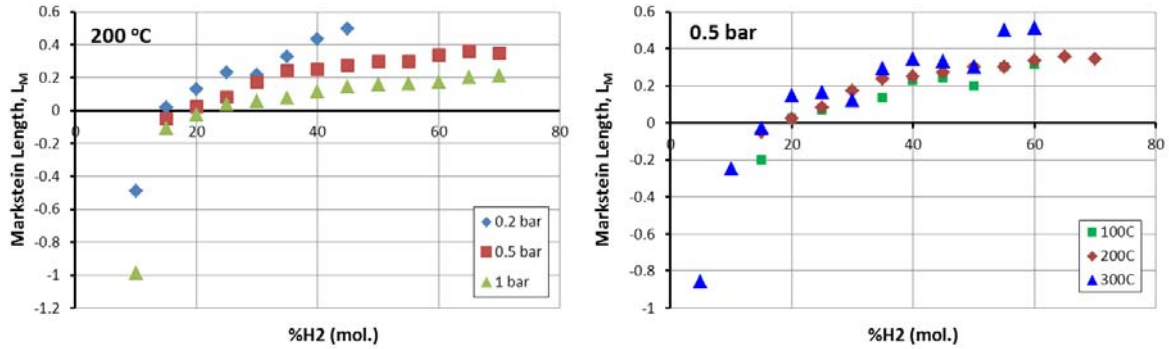


Figure 14. Markstein length vs. H₂ concentration for different initial pressures at 200°C (left) and for different initial temperatures at $p_0 = 0.5$ bar (right).

5.0 CONCLUSIONS

Two methods to obtain flame speeds were presented: the pressure method and an optical method based on a high speed shadow video technique. Free of stretch laminar burning velocities for hydrogen-air mixtures within the flammability limits were determined for temperatures of 20, 100, 200 and 300 °C at reduced pressures from 200 to 1000 mbar.

An overall reaction order n as function of hydrogen concentration was evaluated. It indicates an effect of initial pressure on the laminar burning velocity. It explains that for very lean ($< 20\% \text{H}_2$) and very rich ($> 65\text{--}80\% \text{H}_2$) mixtures the initial pressure has a suppressing effect on the laminar flame velocity ($n < 2$).

It was found that the threshold positive-negative Markstein number L_M shifts from very lean to higher hydrogen concentrations with reduced pressures. A positive L_M at lower pressure, for instance, leads to changing of very fine cellular flame structure of 15 %H₂-air mixture at 1 bar ($L_M < 0$) to smooth flame surface at 200 mbar ($L_M > 0$).

Numerical calculations of laminar flame velocities using Cantera code with two different H/O chemical kinetics mechanisms were performed. Experimental validation of the numerical code showed, that better agreement gives Mueller H/O mechanism than Lutz scheme. Nevertheless, in the range of middle hydrogen concentrations both mechanisms give overestimated calculated laminar flame speed compared to experimental data.

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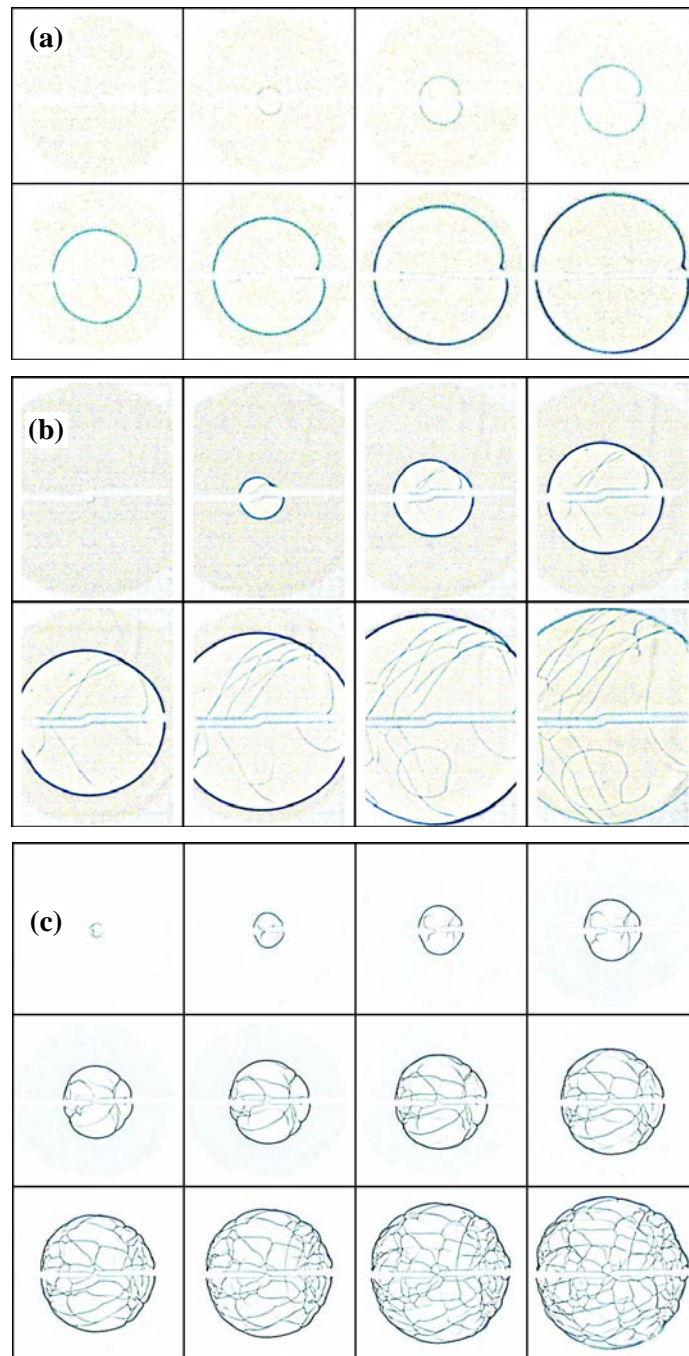


Figure 15. Cellular flame structure for 15 %H₂-air at different initial pressures and temperatures: (a) 200°C, 200 mbar; (b) 300°C, 1000 mbar; (c) 20°C, 1000 mbar

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