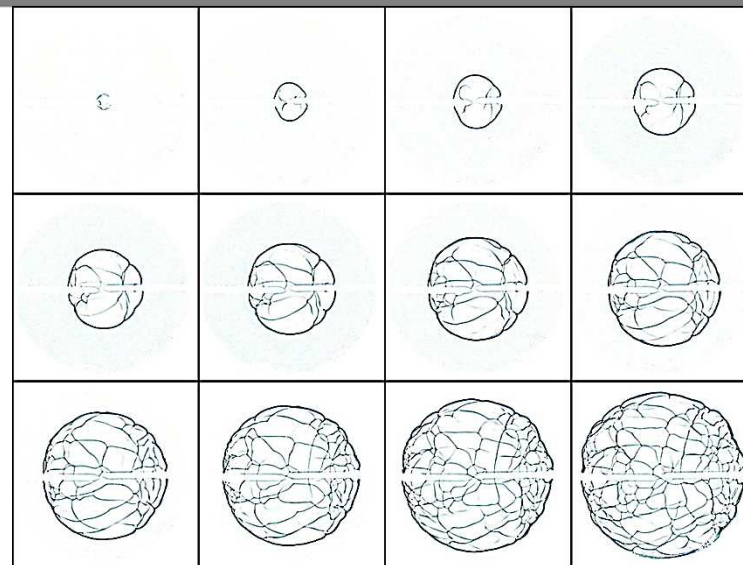# Effect of Temperature on Laminar Flame Velocity for Hydrogen-Air Mixtures at Reduced Pressures
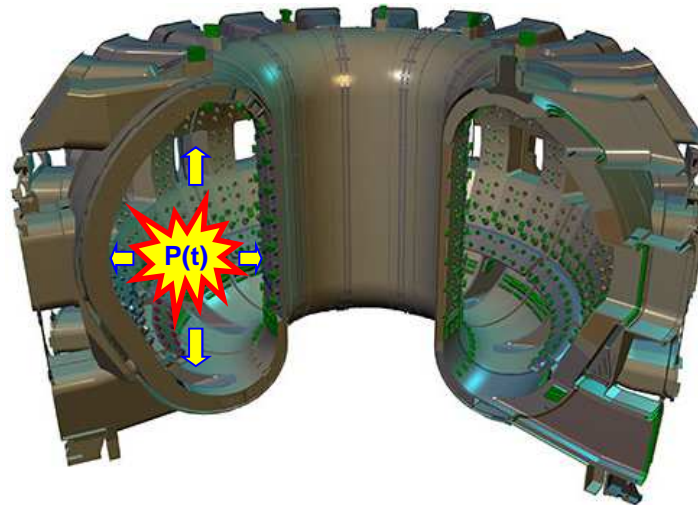
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Institute for Nuclear and Energy Technologies

www.kit.edu

Background

The problem is related to the hydrogen safety of ITER Vacuum Vessel (VV) during Loss of Vacuum Accident (LOVA).


- Hydrogen is produced due to reaction Be + steam and also released from cryo-pumps due to Loss of Cooling Accident (LOCA)
- LOVA scenario: 1-4-20 kg hydrogen inventory + air ingress from ambient atmosphere ➜
  - hydrogen concentration changes from 100 to 1% H2 in air
  - pressure changes from 10 mbar to 1 bar
  - temperature varies from cryogenic to 400C
- Ignition of the mixture may lead to different flame propagation regimes inside the ITER VV
- The problem is to evaluate the combustion properties at reduced pressures.

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VV integrity under combustion pressure loads

**Average hydrogen concentration**
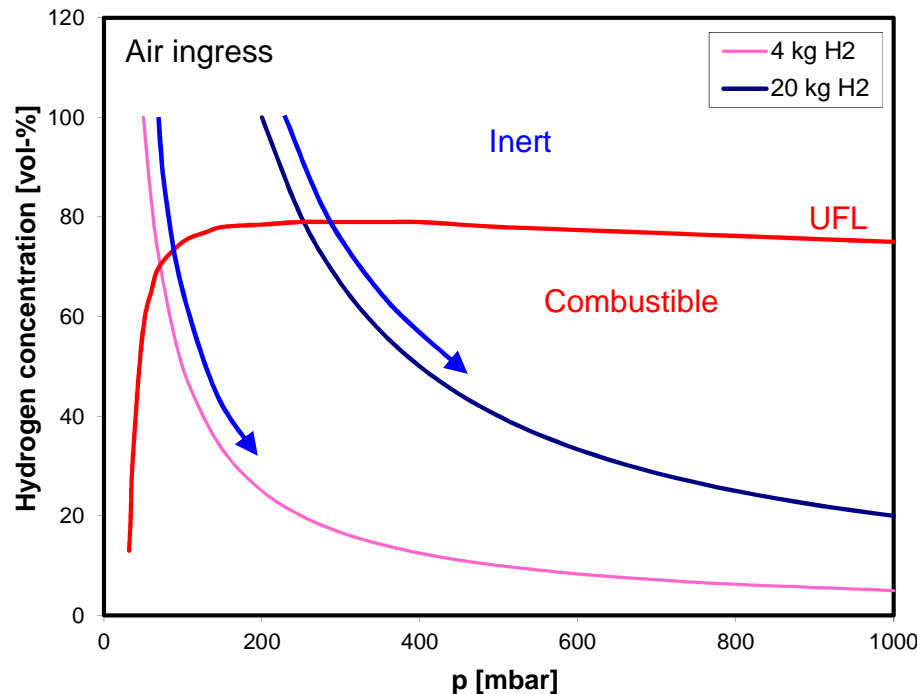
Maximum combustion pressure
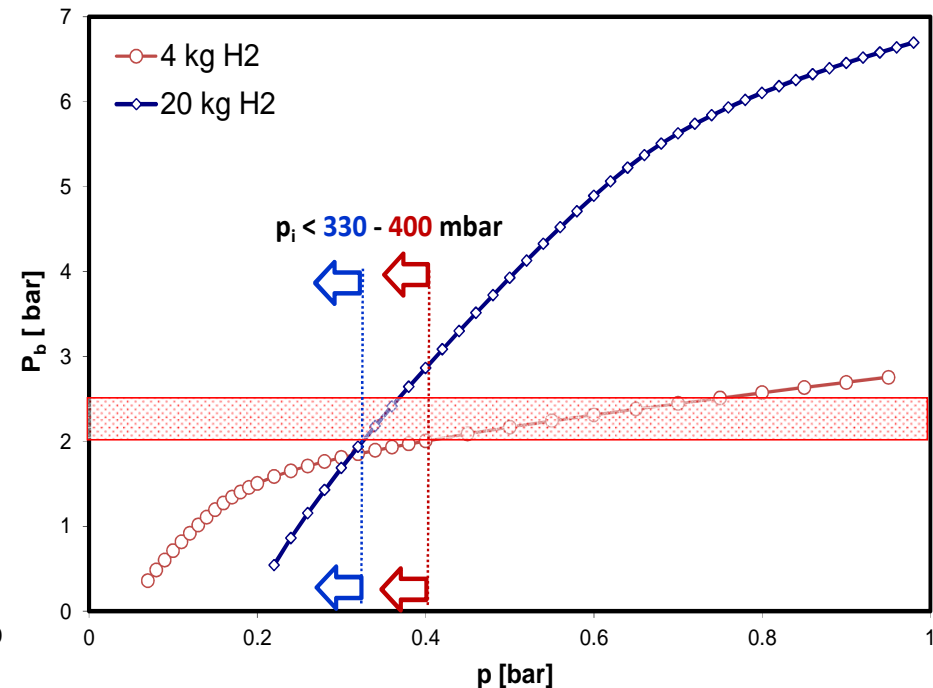
- Mixture inside the VV becomes burnable after hydrogen concentration reaches the upper flammability limit (UFL)

- Maximum adiabatic combustion pressure may exceed design pressure of ITER VV
  $P_{max}= 2$ bar at the pressure of ignition $P_0 > 350$ mbar (in the case of laminar deflagration)

- In case of sonic flame or detonations the peak pressure may exceed design pressure of ITER VV at an initial pressure $P_0 \sim 275$ mbar (just at the flammability limit)

Flammability limits of hydrogen-air mixtures

P = 1 bar; T = var
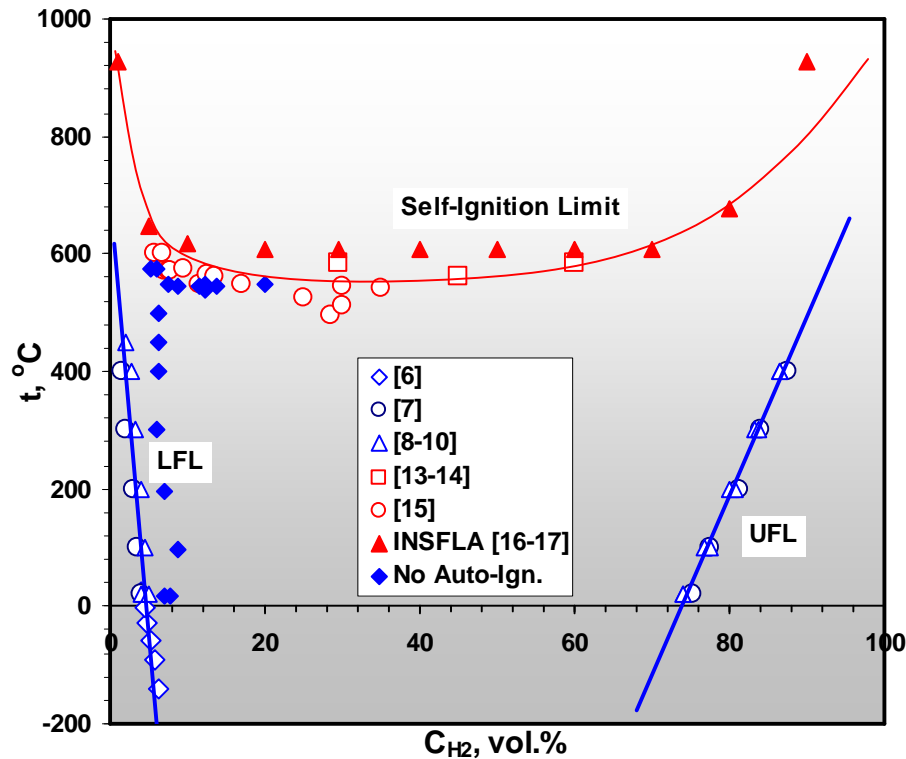
**P = var; T = 293 K**
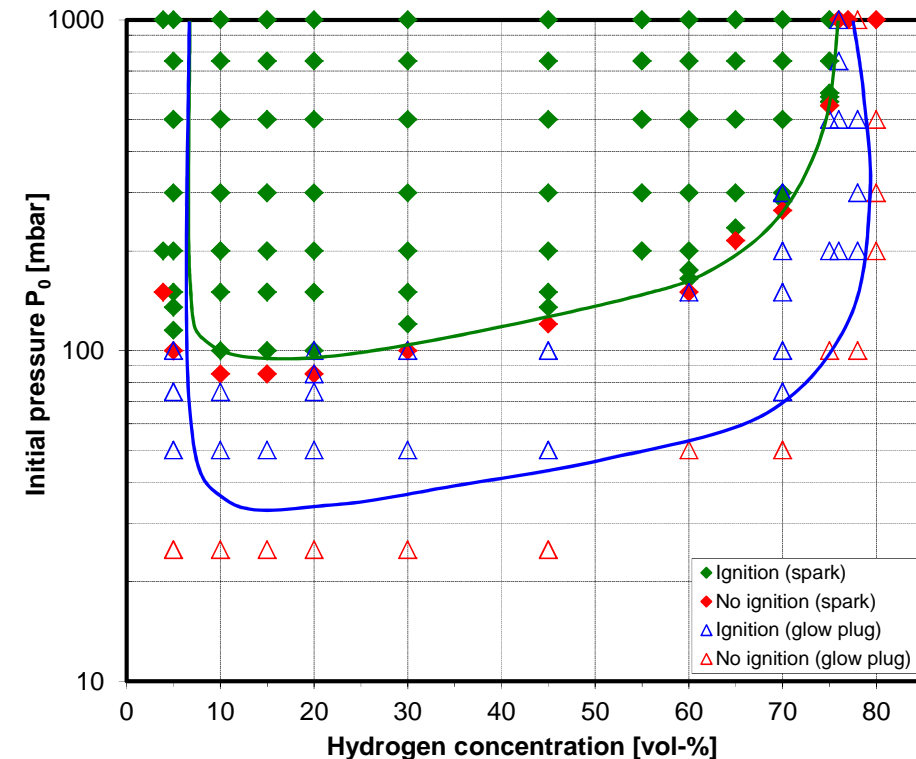
- Promoting effect of the temperature on the flammability limits
- The Lower Flammability Limit (LFL) is not sensitive to the initial pressure. It remains at about 4%H2 in air up to 50 mbar as it is at atmospheric pressure.
- The UFL extends from 75%H2 at 1000 mbar to 80%H2 at pressures 200-500 mbar.
- Below 200 mbar the UFL reduces from 80%H2 to 50%H2 at a pressure of 50 mbar
- Hydrogen-air mixture at initial pressure lower than 25 mbar could not be ignited with an ignition energy up to 4.3J.

9.2013

Minimum ignition energy
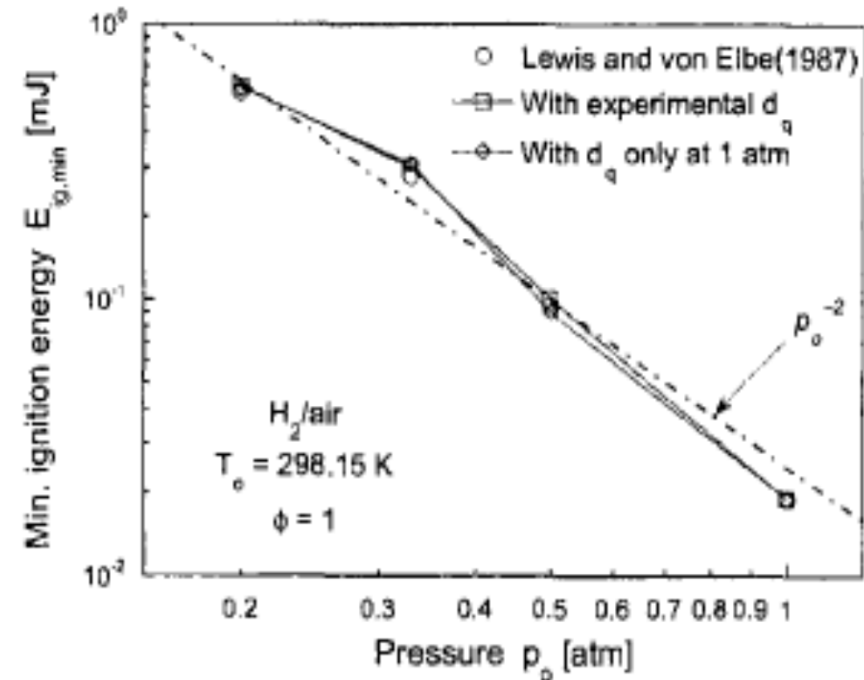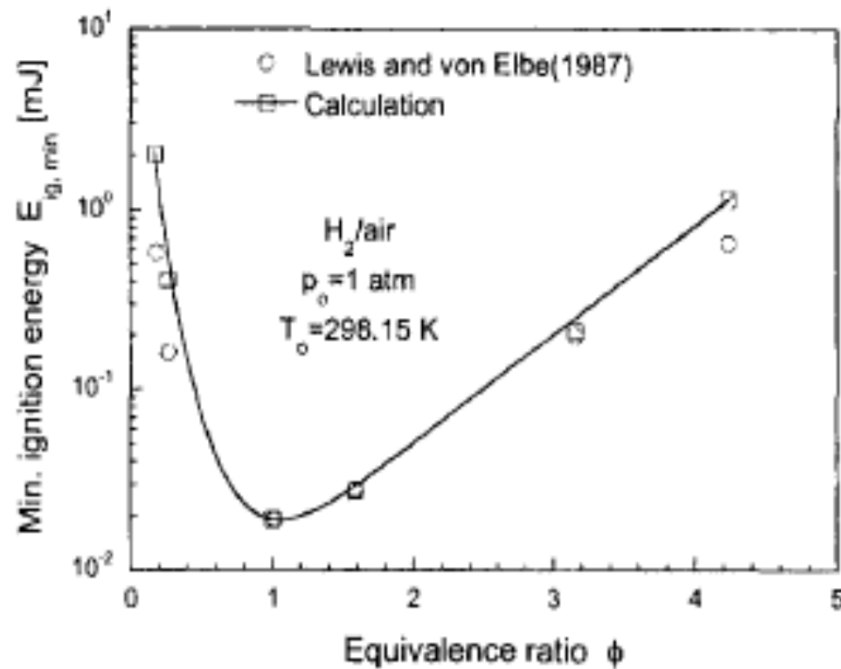
- Minimum ignition energy increases with pressure ($\sim p^{-2}$)
- Very rapid minimum ignition energy increase close to the flammability limits

Experimental results on ignition energy
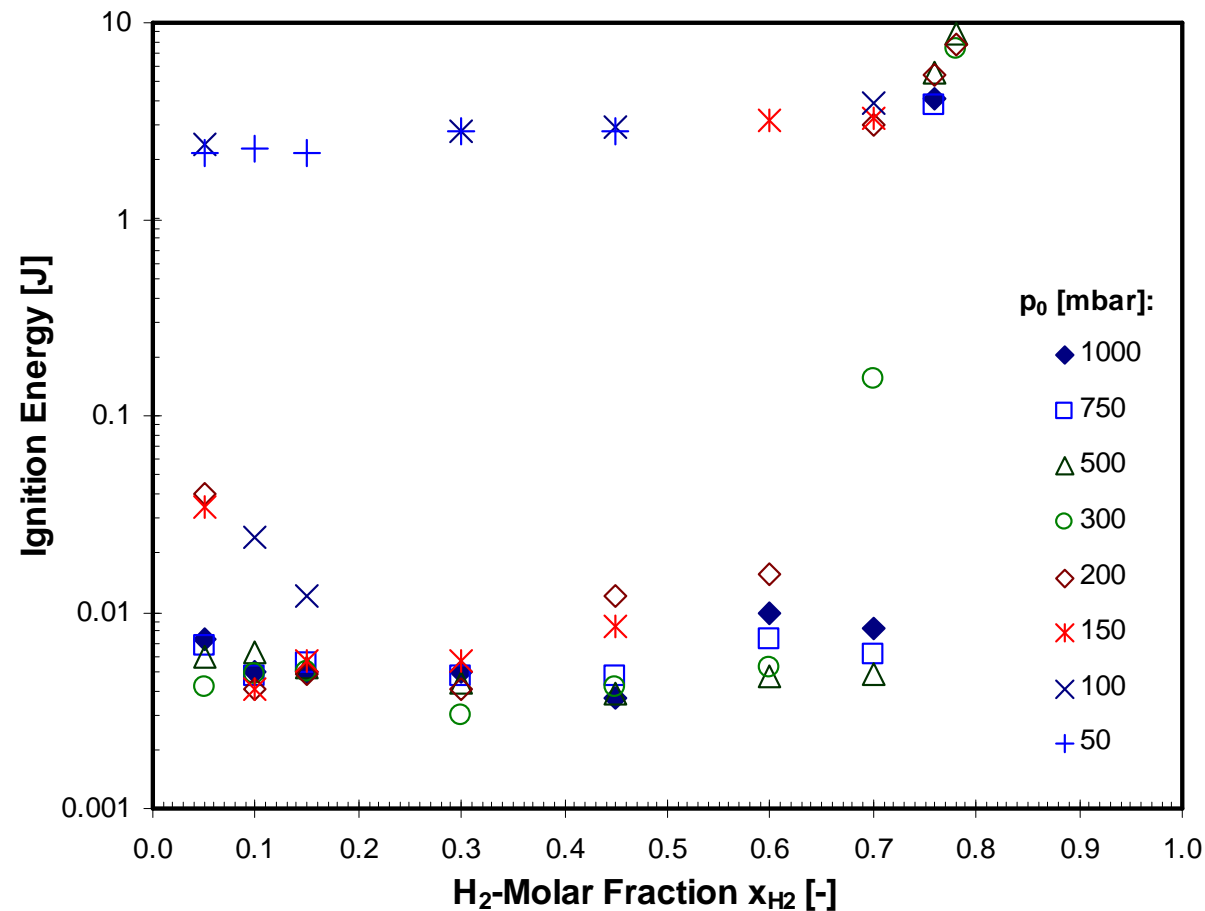
- Ignition energy increases up to several Joules with pressure decrease from 1 bar to 50 mbar
- Close to the flammability limit the ignition energy increases up to 7.5 J from the UFL side

Why laminar flame speed data are important?

- Used for reaction mechanism and numerical code validation
- Fundamental property of combustible mixtures
- S_L is an overall characteristic of reactivity:

Evaluation of overall kinetic characteristics of RG mixtures from data on LBV (Frank-Kamenetskii approach)

$$k_0 e^{-\frac{E_a}{T_b}} = (\sigma S_L)^2 \frac{(T_b - T_u)^{n+1}\left(\frac{T_u}{T_0}\right)^{n-1} E_a^{n+1}}{\nu_1 X_1^{n_1-1} X_2^{n_2}\left(\frac{p}{p_0}\right)^{n-1} \chi(T_b) Le^n T_b^{2(n+1)}\Gamma_{n+1}}$$

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

- overall energy activation E_a:

$$E_a = \frac{\Delta[(2n+2)\ln T_b + (n-1)\ln p - (n+1)\ln(T_b - T_u) + \ln(\chi) - 2\ln(\sigma S_l)]}{\Delta\left(\frac{1}{T_b}\right)}$$

$$E_a = -2\frac{\partial \ln(\sigma S_l)}{\partial(1/T_b)}$$

- reaction order n (sensitivity to the pressure):

$$n = 2\frac{\partial \ln(S_l)}{\partial \ln(p)} + 2$$

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

- laminar flame thickness δ is a scaling factor:

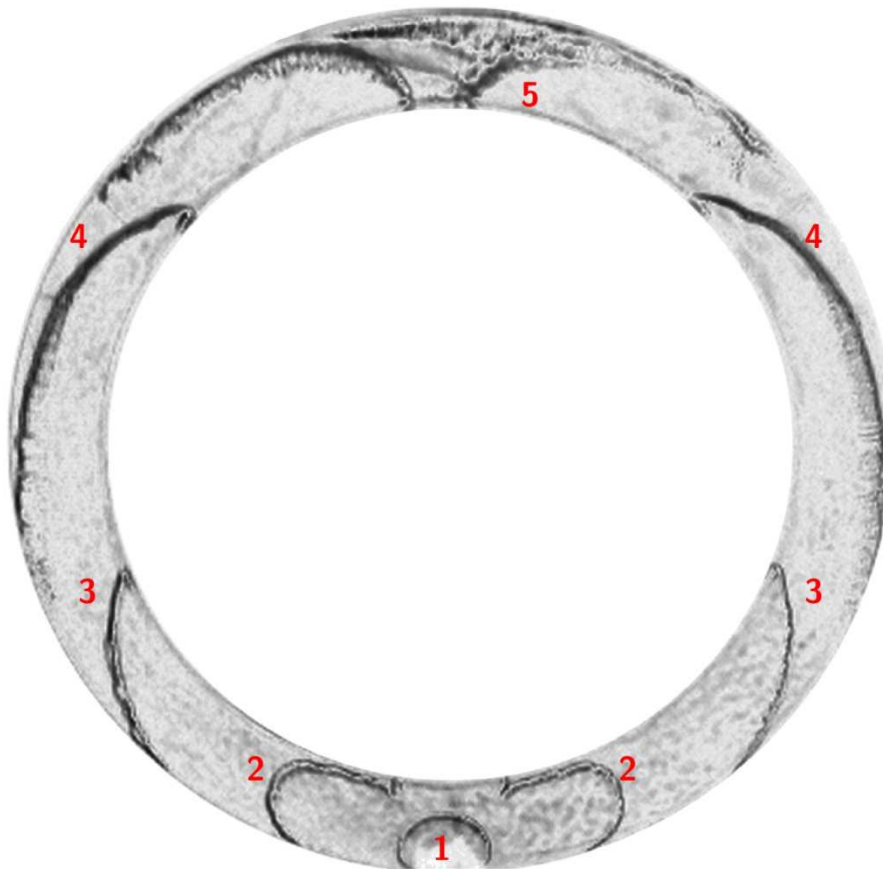$$\delta = \frac{(T_b - T_u)}{\left(\frac{dT}{dx}\right)_{max}}$$

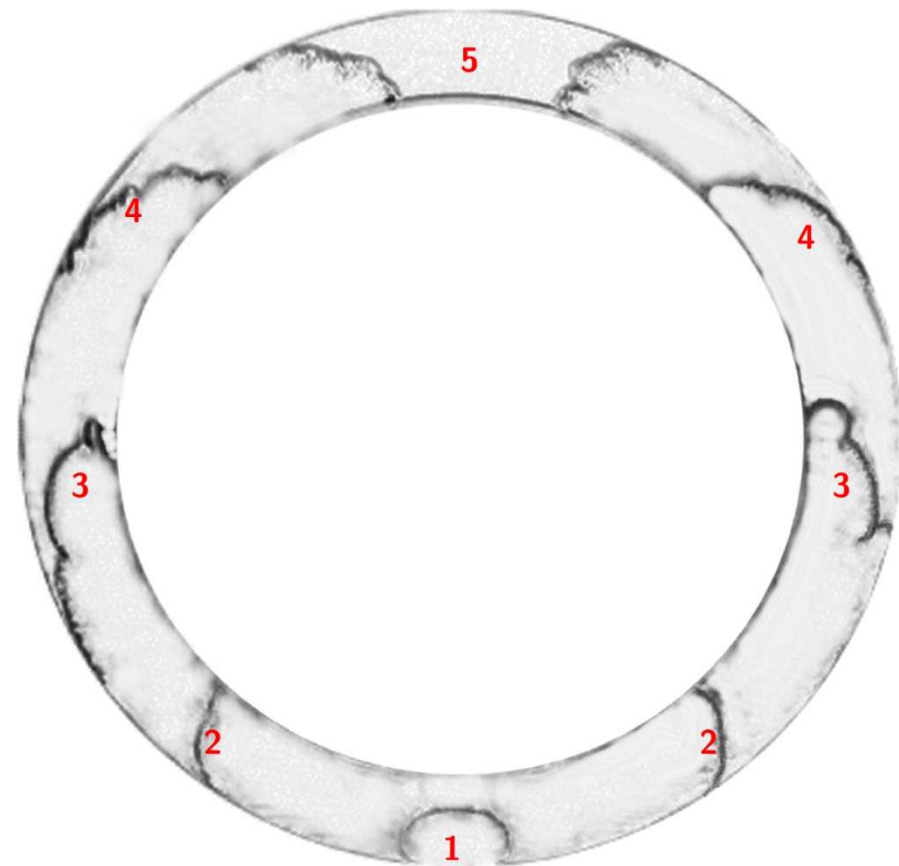- Markstein number (length) Ma (L_b):

$$Ma_b = L_b / \delta$$

$$Ma_b = (\sigma/(\sigma-1))\cdot(\ln\sigma + (Ze(Le-1)/2/(\sigma-1))\cdot \int_0^{\sigma-1}(\ln(1+x)/x)dx)$$

Flame propagation in torus geometry

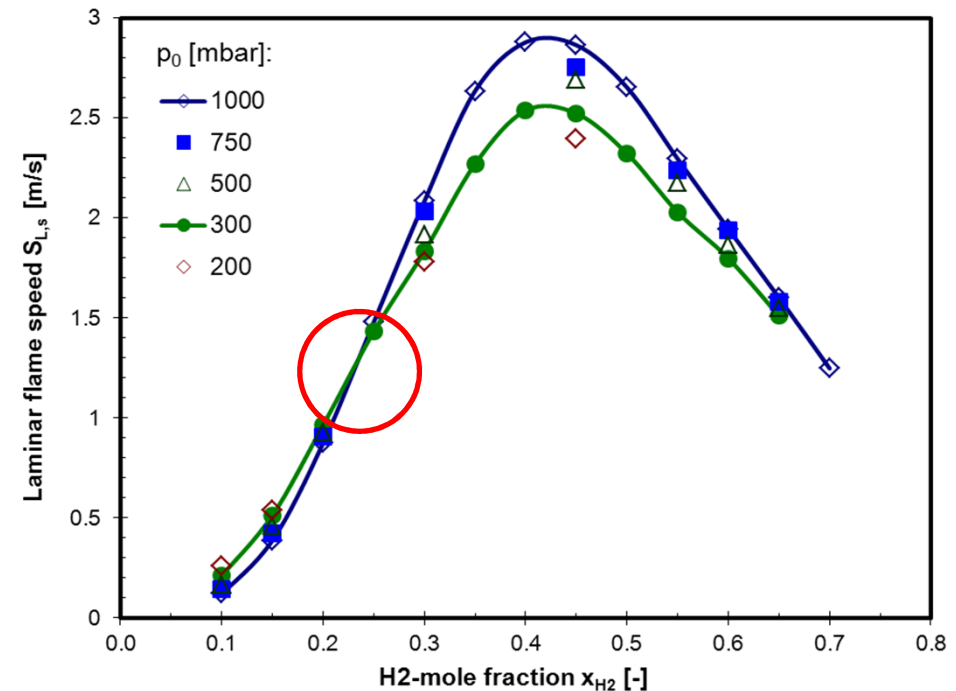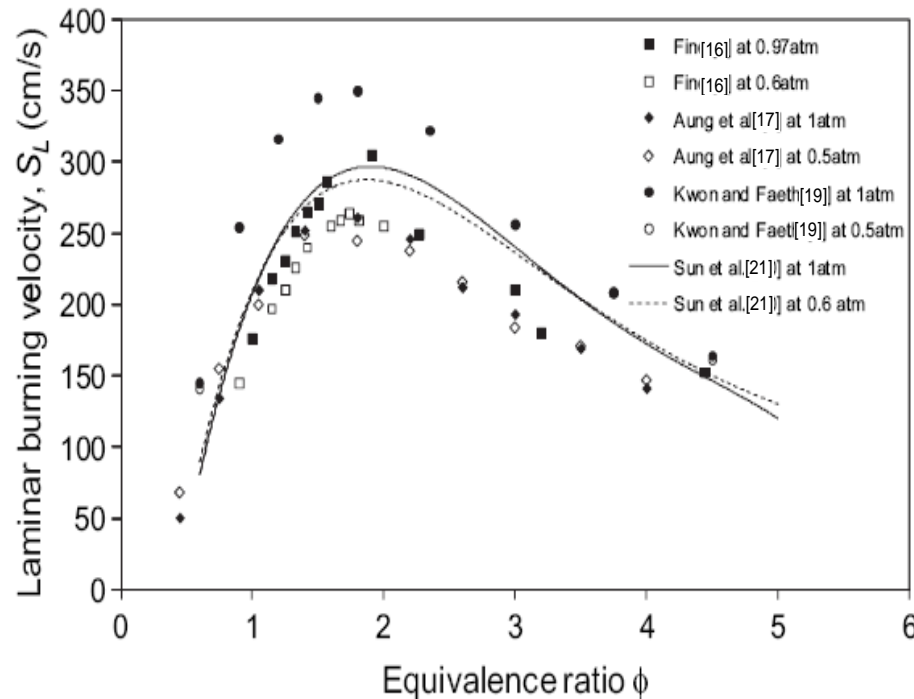**Effects of torus curvature and flame curvature on flame dynamics**

50 Vol.-% H$_2$

80 Vol.-% H$_2$

# Experimental data on laminar burning velocities of H_2-air mixtures

- All reference data on hydrogen-air laminar flame speed at reduced pressure obtained at ambient temperature. The lack of data at elevated temperatures

- The lowest initial pressure was reached is 200 mbar

- Pressure dependence of laminar velocity depends on reaction order: $S_L(P) \propto S_L(P_0) \cdot \left(\dfrac{P}{P_0} \right)^{\frac{n}{2}-2}$
  $S_L \uparrow$, $P \uparrow$ (n :> 2); $S_L \downarrow$, $P \downarrow$ (n :< 2);

- The threshold between positive and negative effects of initial pressure lies at 15-20%H2

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 $^{\circ}$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

Laminar flame velocity calculations
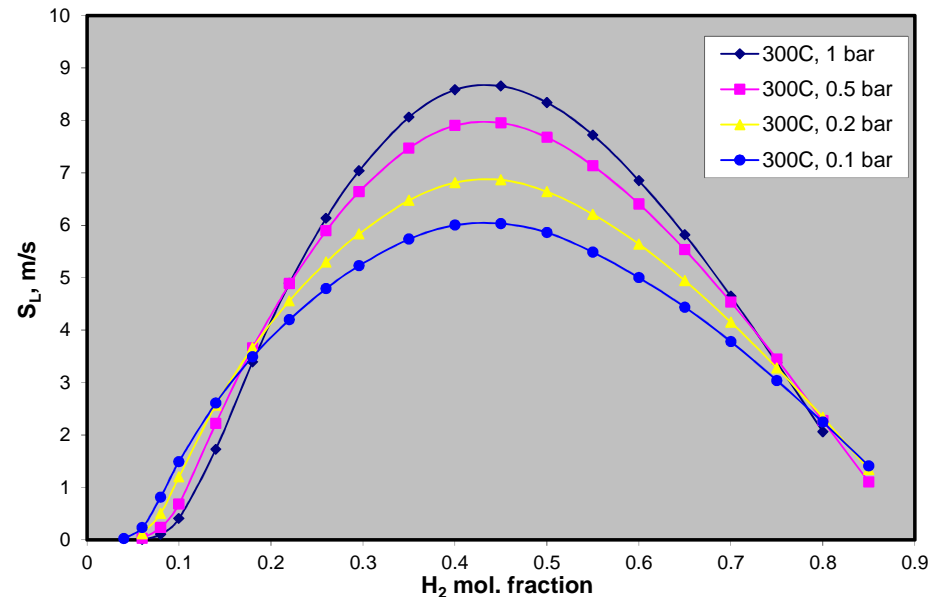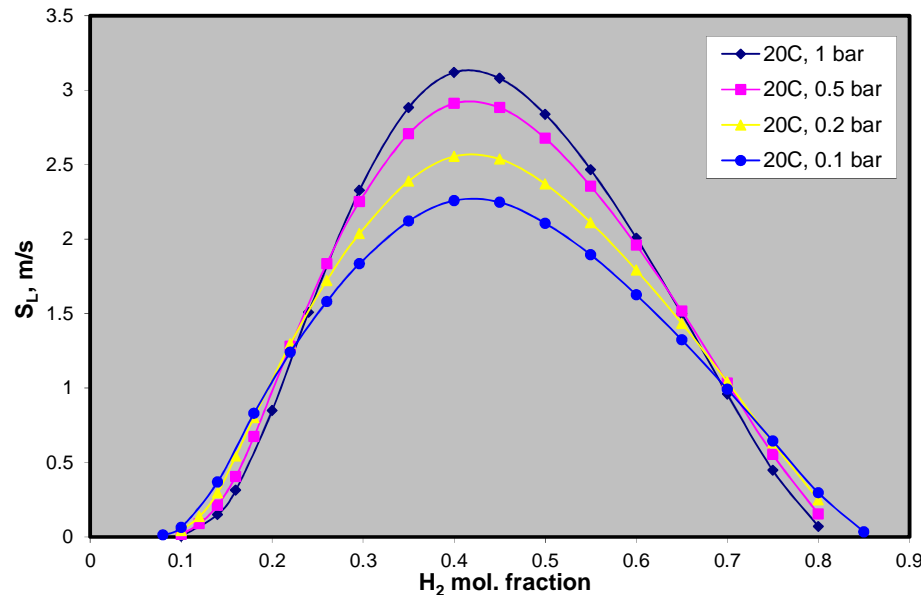
- Numerical calculations of laminar burning velocities were obtained using Cantera code for unstretched (plane) flames.

- Different H/O reaction mechanisms (Lutz, Mueller and Keromnes) were used in this work.

- Maximum laminar flame velocity increases almost 3 times with temperature increase from 20 to 300C

- A threshold between positive – negative pressure effect on flame velocity is located at 20 and 67-77%H2

# Laminar flame velocity calculations. A comparison
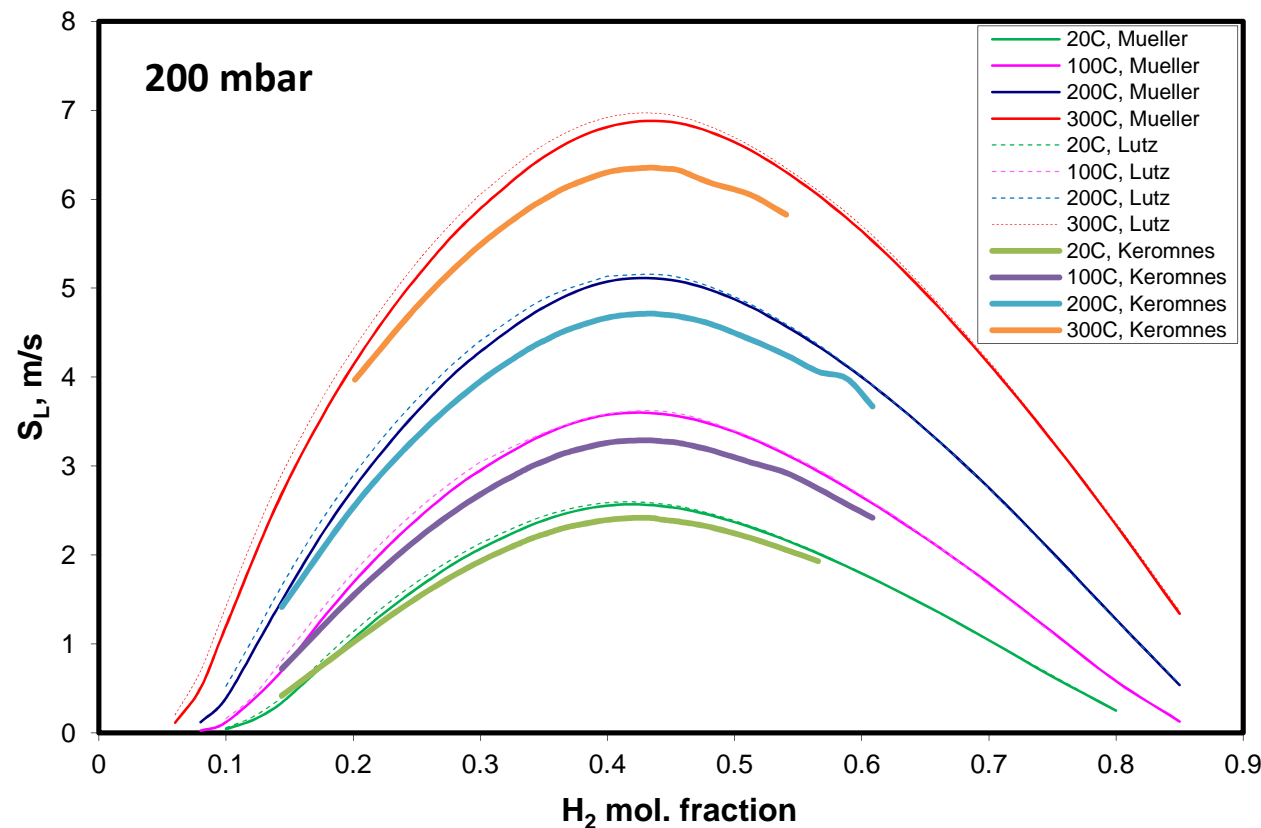
- Different H/O reaction mechanisms (Lutz, Mueller and Keromnes) were used in this work.
- All the mechanisms give quite similar data on laminar flame velocity with a difference that Lutz data a bit higher and Keromnes data are the lowest, especially at higher temperatures.
- Keromnes and Mueller mechanisms as the most suitable for sub-atmospheric pressures were chosen for further data analysis

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Overall reaction order

- According to Zeldovich-Frank-Kamenetskii theory the pressure dependence of laminar flame speed can be written $S_L \sim p_0{}^\beta$, where $\beta = n/2-1$. Pressure exponent is negative and the initial pressure has suppressing effect on the laminar flame speed if reaction order n < 2.

- The overall reaction order n is a parameter responsible for the pressure dependence of laminar velocity. The overall reaction order n was obtained by changing the initial pressure p_0.
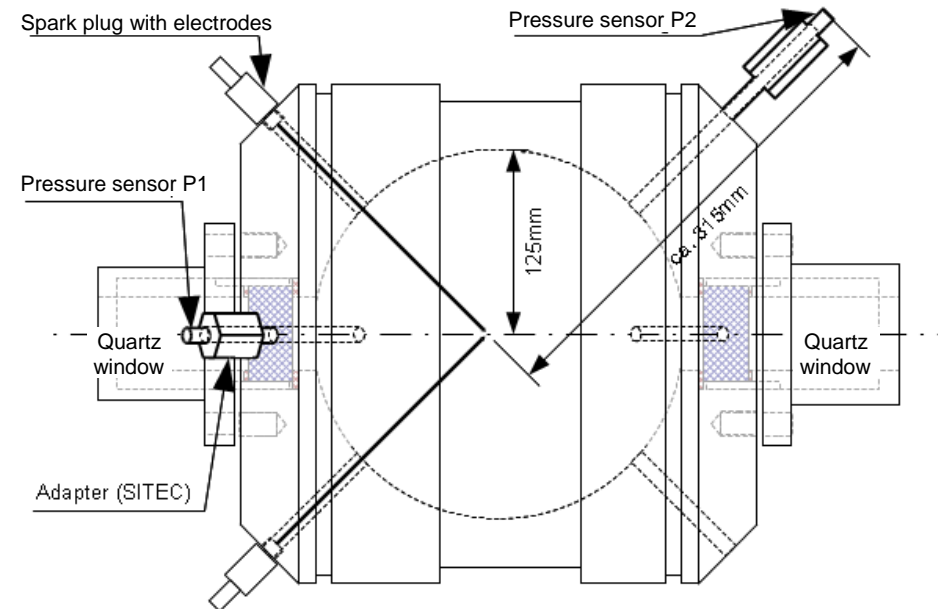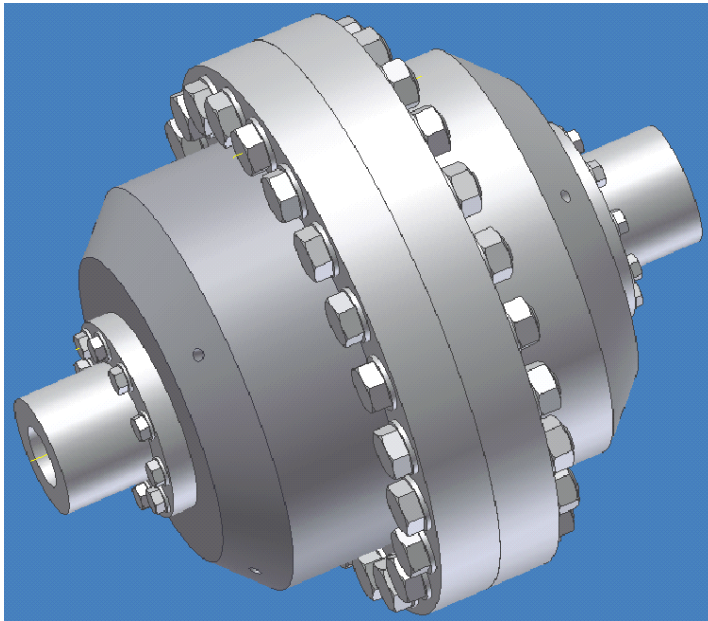
$$n = 2\frac{\partial \ln(S_L)}{\partial \ln(p)} + 2$$
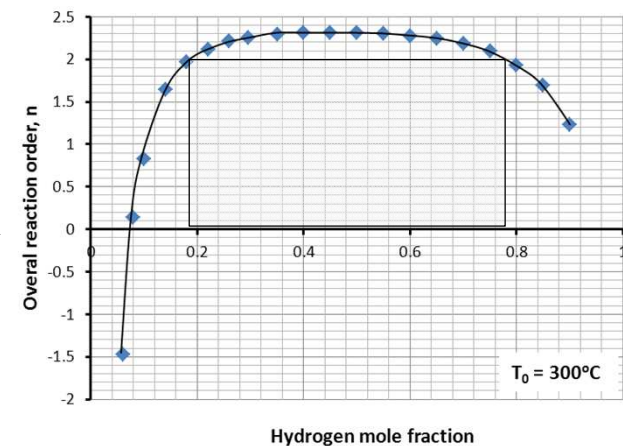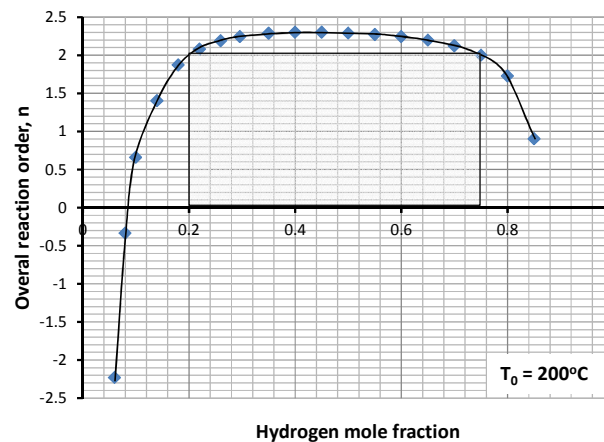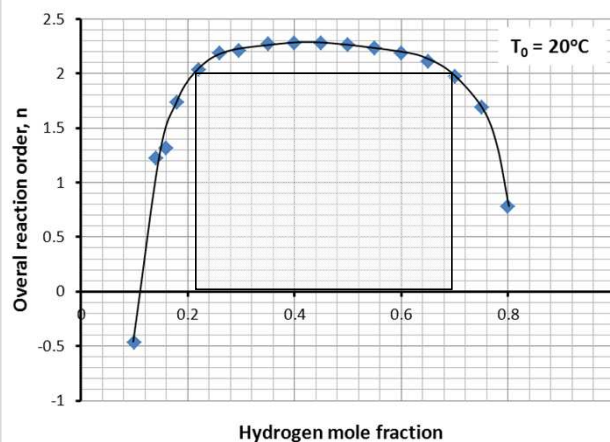
- Overall reaction order vs. H2 concentration was calculated at different temperatures.

- The overall reaction order increases at higher temperatures in the vicinity of lower and upper flammability limits

Dr. Mike Kuznetsov, IKET

Experimental facility

Spherical Explosion Bomb


- Stainless steel vessel: (WN 1.4571/SS 316L)
- Size of internal volume: D = 250 mm (V=8.2 dm^3)
- Wall thickness: t > 34 mm
- Maximum design pressure: 800 bar
- Spark plug E =0.3-1000 mJ
- Glow plug E =200-4300 mJ

- 2 piezoresistive transducers
- Glass windows + high speed camera (250000 fr/s)
- Static pressure sensors
- Data acquisition system: slow/fast ADC controllers (1 MHz sampling rate)
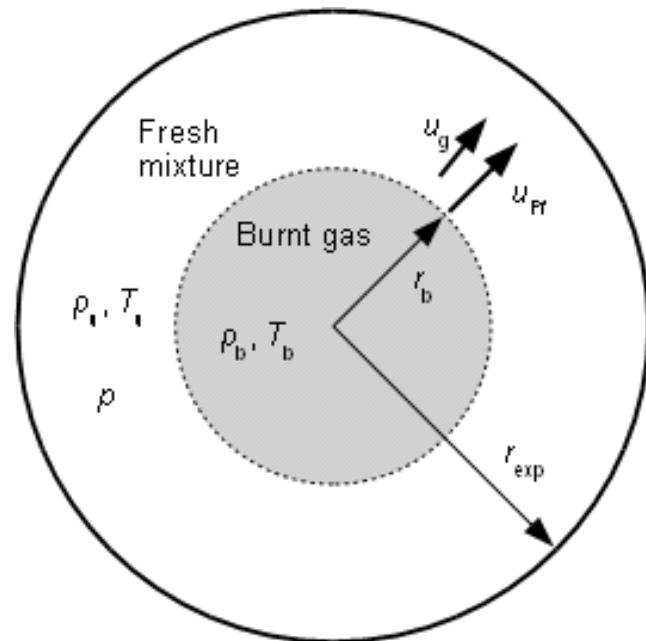
Laminar burning velocity. Schlieren technique

60%H$_2$-air

15%H$_2$-air

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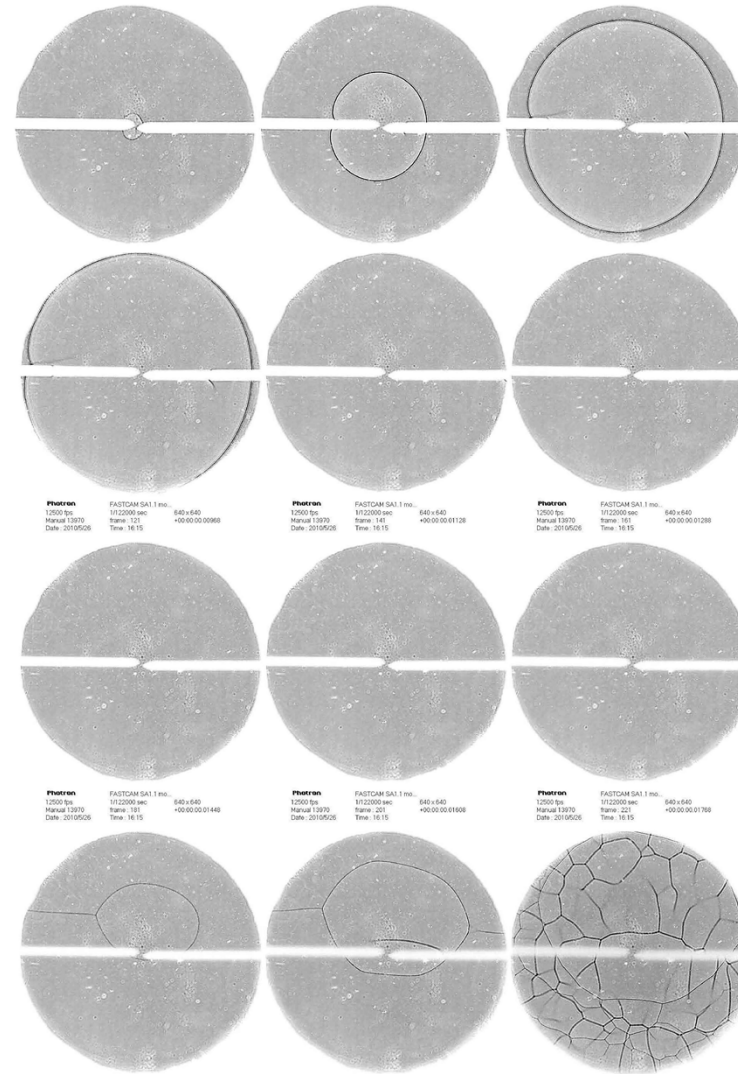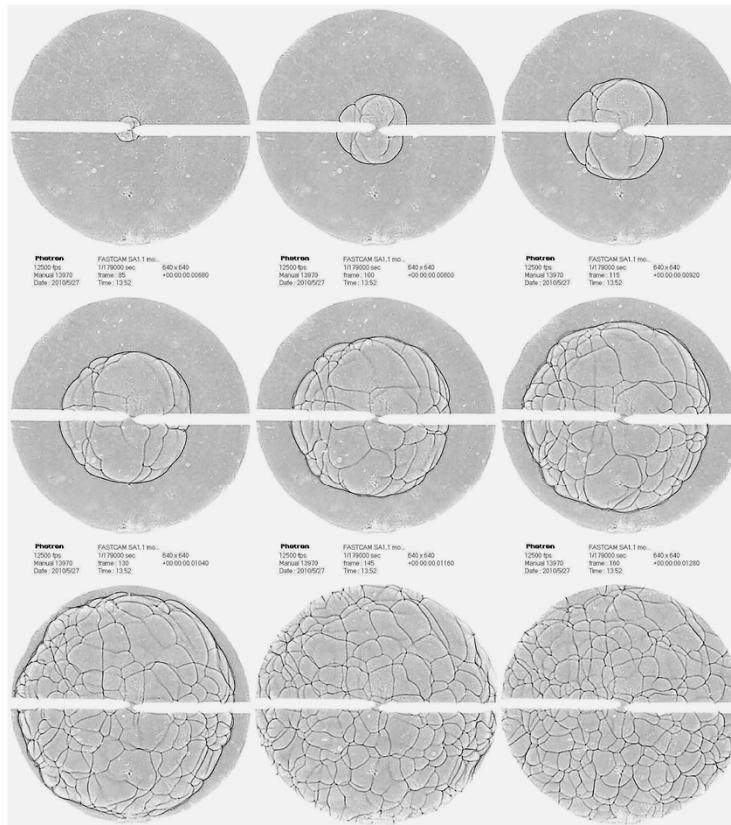
Laminar flame velocity evaluation

- Effect of flame curvature on the laminar flame speed
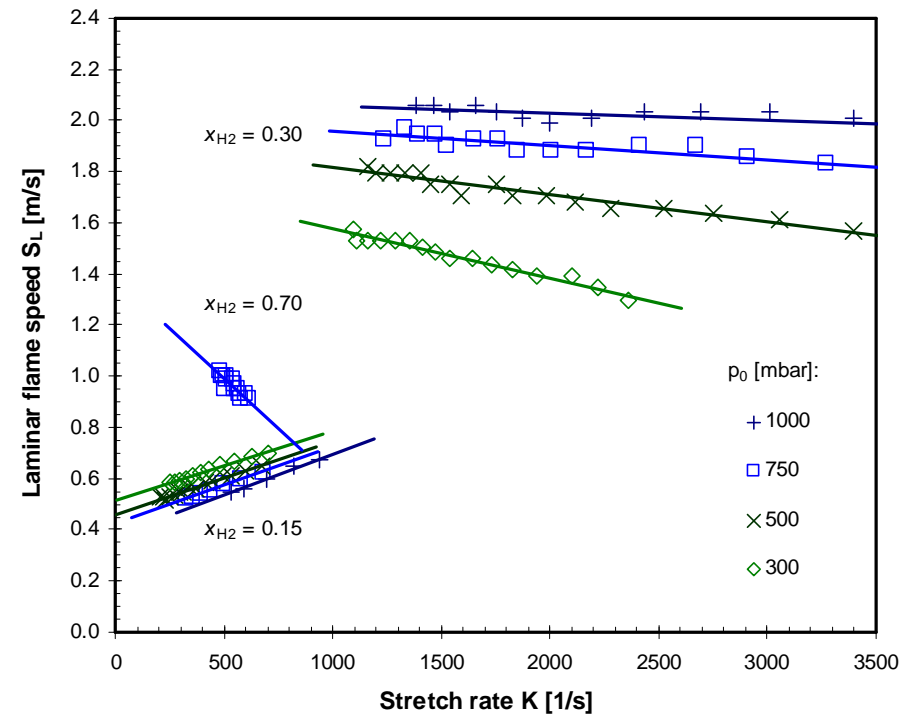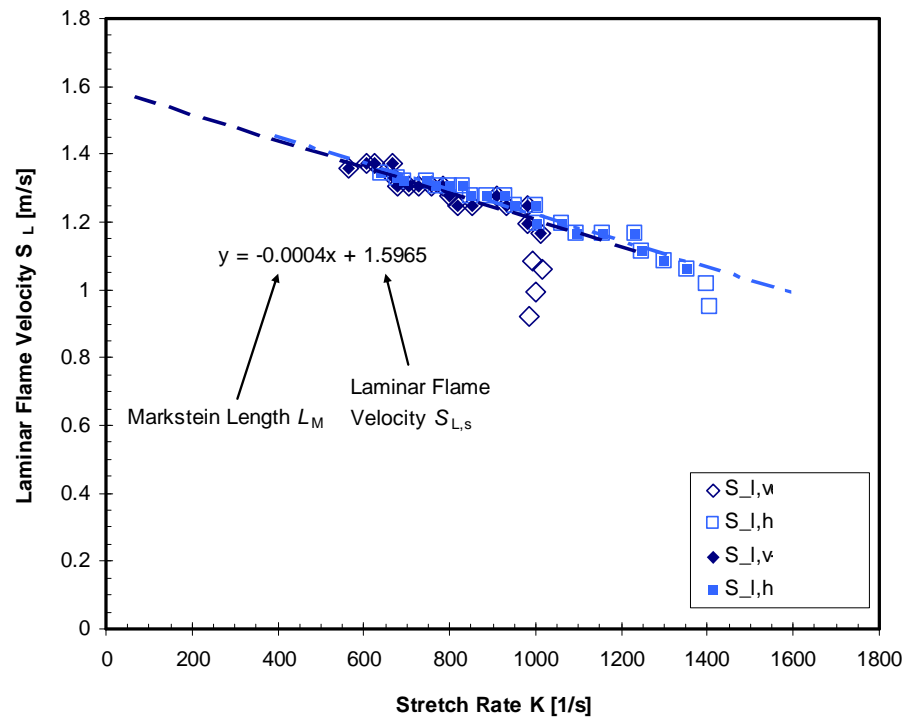
$$S_L = S_{L,s} - L_M \cdot K$$

where L_M is the Markstein length; strain rate

$$K = \frac{1}{A}\frac{dA}{dt} = \frac{2}{r_b}\frac{dr_b}{dt} \quad \text{(for spherical case)}$$

- Positive or negative influence of the flame curvature (or sphere radius) on the laminar velocity.

- A positive Markstein length 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.

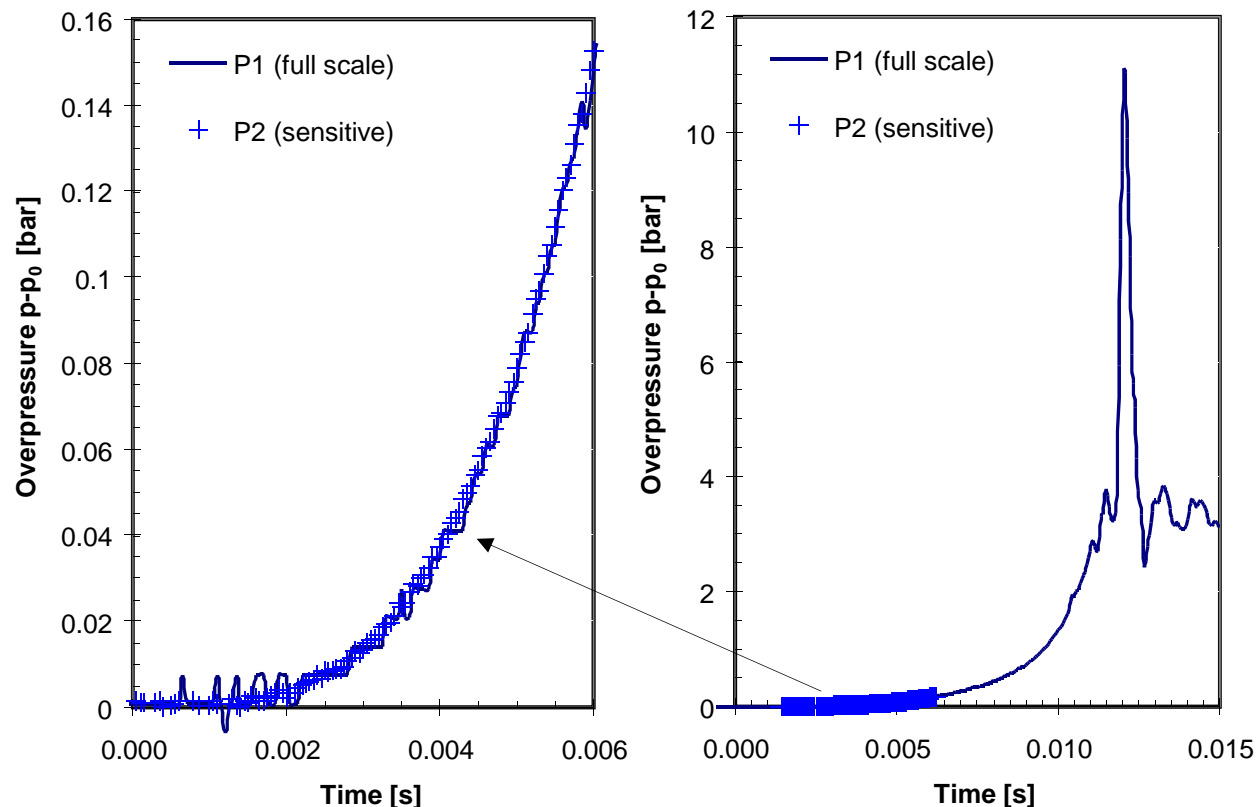# Schlieren technique. Data processing

65%H$_2$-air, 750 mbar

$$S_L = S_{L,s} - L_M \cdot K$$

Laminar burning velocity

Spherical bomb method. Typical pressure records

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

$$S_L(t) = \frac{r_{\exp}}{3} \frac{1}{p_e - p_0} \left(\frac{p_0}{p} \right)^{1/\gamma} \left[1 - \left(\frac{p_0}{p} \right)^{1/\gamma} \frac{p_e - p}{p_e - p_0} \right]^{-2/3} \frac{dp}{dt}$$
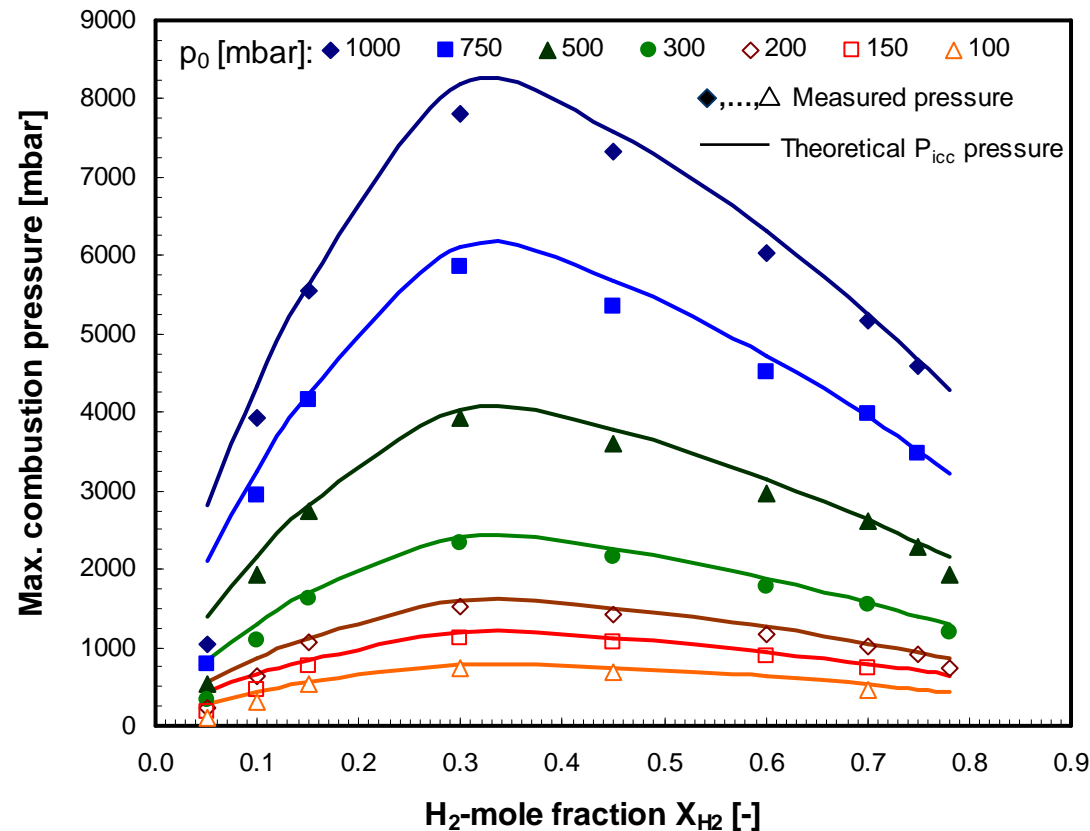
S_L - stretched laminar flame velocity

p_0, p_e – initial and final pressure;

R – bomb radius;

r_b – burned gas radius

Only initial part of pressure records ($p < 0.02 P_e$) was used

Pressure method. Maximum combustion pressure

- Good agreement between theoretical P_{icc} pressure and measured maximum combustion pressure
- Only approaching to the lower flammability limit the difference is rather big because flame instability and incomplete combustion

Laminar flame velocity evaluation
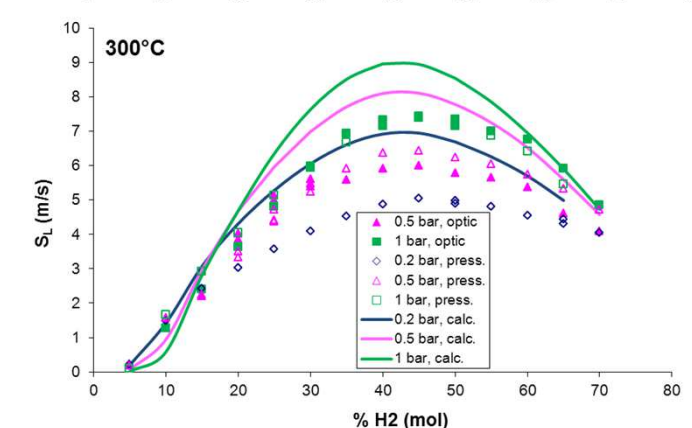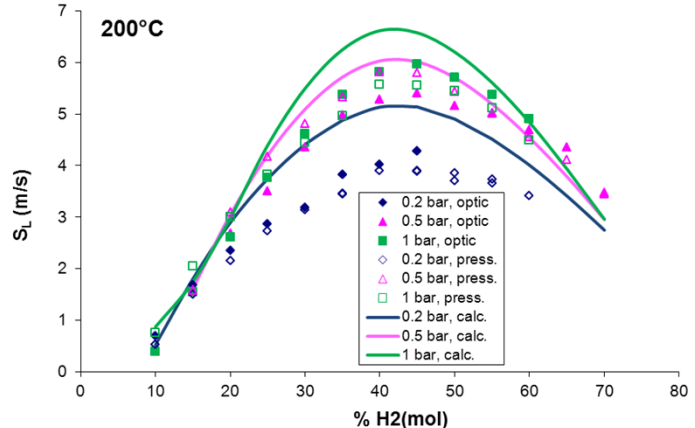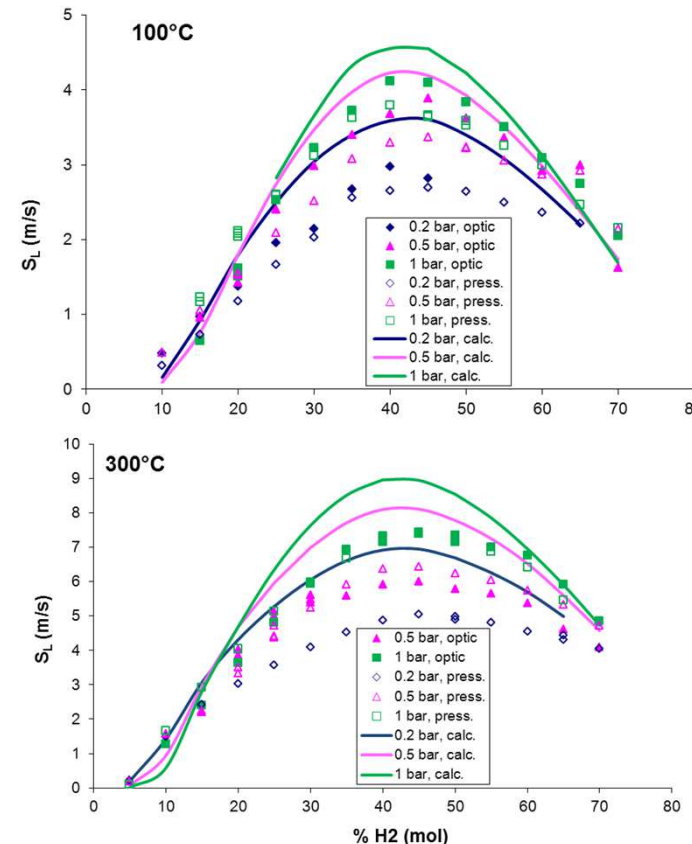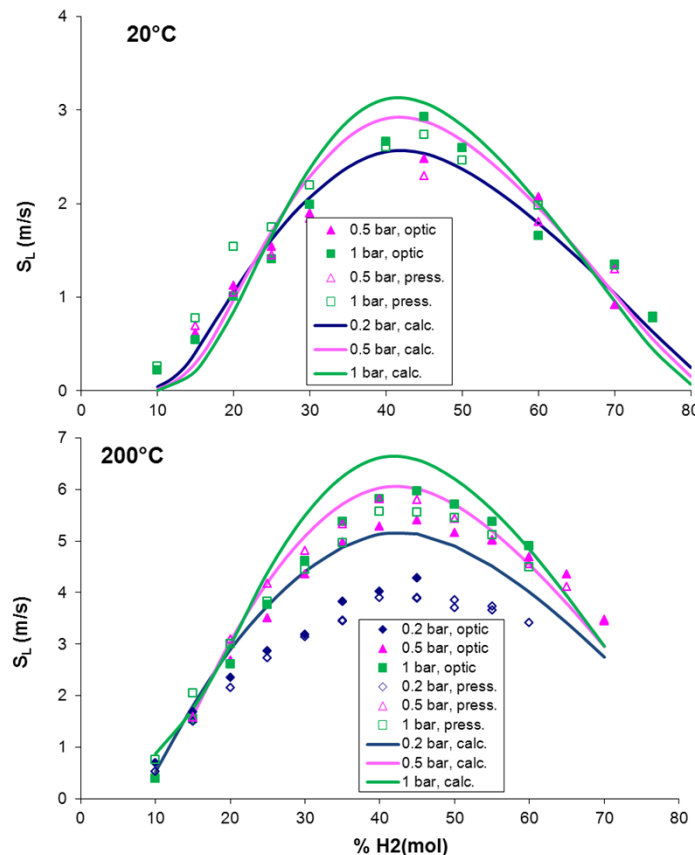
- Very good consistency of optical and pressure measurements in current work

- The lower the initial temperature the better is agreement of calculations to experimental data

- Even one of the best Mueller mechanism is quite close or underestimated the experimental values of SL at lean and rich hydrogen concentrations.

- In the range of middle hydrogen concentrations the calculations with all mechanisms overestimated the experimental values of SL.

Markstein length
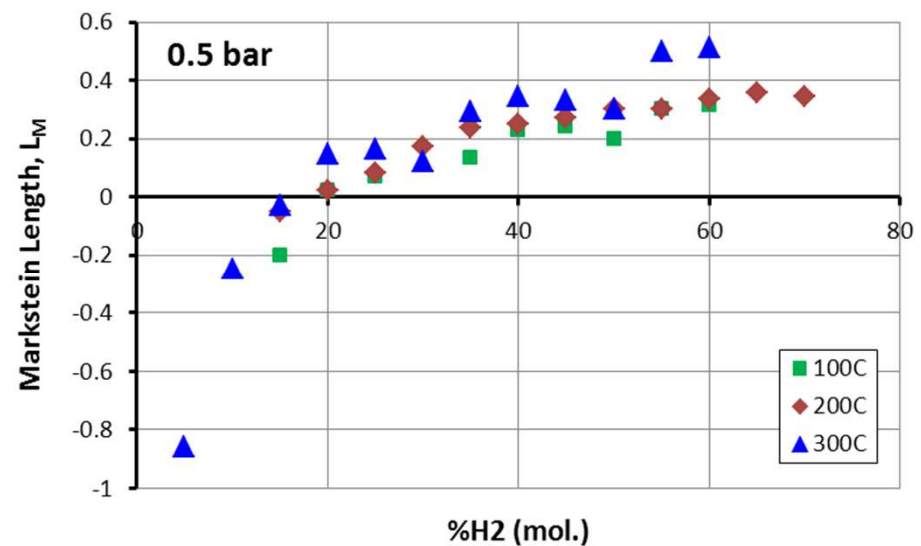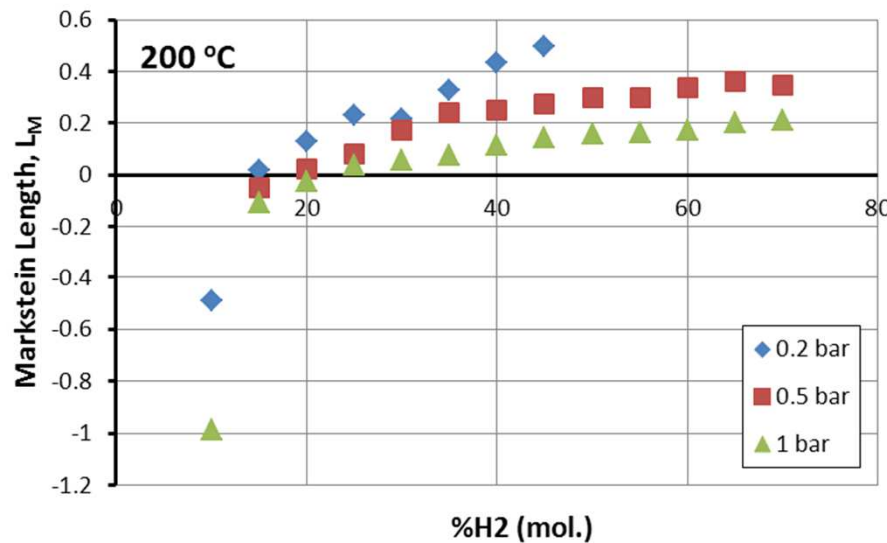
- The influence of flame front stretch on laminar flame speed can be specified by Markstein length L_M: $S_L = S_{L,s} - L_M K,$ where $K = 1/A*dA/dt$.

- For all the concentrations, the Markstein length increases progressively with decreasing initial pressure. Also, the concentration of fuel has a great influence on the effect of the flame front curvature. Lean mixtures with $x_{H2} < 0.2$ have negative Markstein lengths, while for stoichiometric and rich mixtures the positive Markstein lengths was measured.

- For lean mixtures the stretch effects cause an acceleration of the curved flame front to the planar, while for stoichiometric and rich mixtures stretch effect cause a deceleration of the flames.

The effect of Markstein length on flame cellularity

15%H_2-air

200°C, 200 mbar

300°C, 1000 mbar

20°C, 1000 mbar

P↓

T↗
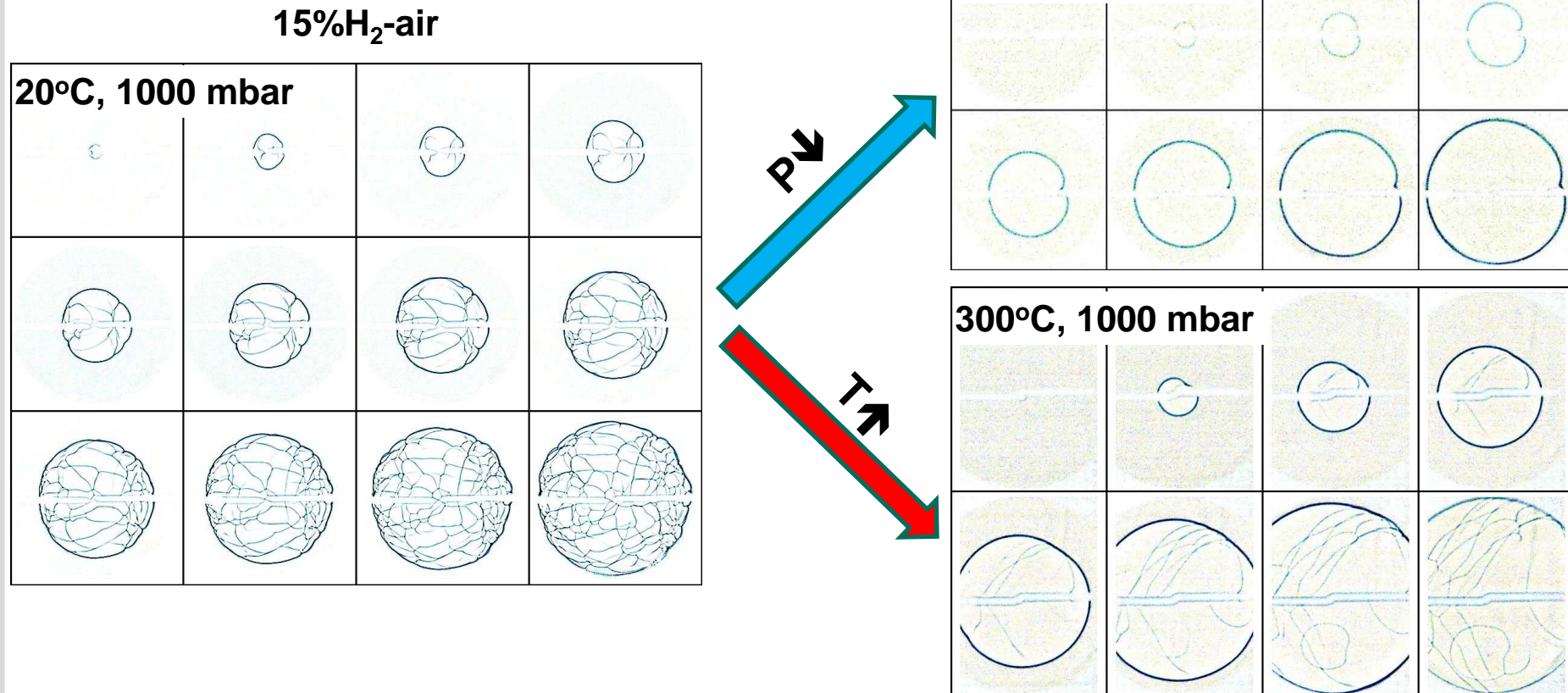
- A positive Markstein length at lower pressure leads to changing of very fine cellular flame structure of 15 %H2-air mixture at 1 bar ($L_M < 0$) to almost smooth flame surface at 200 mbar ($L_M > 0$)

- Effect of elevated temperature is not so significant but, nevertheless, according to the temperature dependence of Markstein number L_M the cellular structure at 300°C becomes larger compared to very fine structure at 20°C

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Conclusions

- **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 laminar burning velocity It shows that for very lean (< 20%H2) and very rich (> 65-80%H2) mixtures the initial pressure has a suppressing effect on the laminar flame velocity (n < 2).**

- **The threshold positive-negative Markstein number LM shifts from very lean to higher hydrogen concentrations with reduced pressures. A positive LM at lower pressure leads to changing of very fine cellular flame structure of 15 %H2-air mixture at 1 bar (LM < 0) to smooth flame at 200 mbar (LM > 0).**

- **Numerical calculations of laminar flame velocities using Cantera code with Keromnes, Mueller and Lutz H/O chemical kinetics mechanisms were performed. With the best agreement of Keromnes and Mueller mechanism, in the range of middle hydrogen concentrations all calculations give overestimated laminar flame speed compared to experimental data**

Thank you!

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