EXPERIMENTAL INVESTIGATION ON THE BURNING BEHAVIOR OF HOMOGENEOUS H2-CO-AIR MIXTURES IN AN OBSTRUCTED, SEMI-CONFINED CHANNEL

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ABSTRACT

In the current work the combustion behavior of hydrogen-carbon monoxide-air mixtures in semi-confined geometries is investigated in a large horizontal channel facility (dimensions 9 m x 3 m x 0.6 m (L x W x H)) as a part of a joint German nuclear safety project. In the channel with evenly distributed obstacles (blockage ratio 50%) and an open to air ground face, homogeneous H2-CO-air mixtures are ignited at one end. The combustion behavior of the mixture is analyzed using the signals of pressure sensors, modified thermocouples and ionization probes for flame front detection that are distributed along the channel ceiling. In the experiments various fuel concentrations (cH2 + cCO = 14 to 22 Vol%) with different H2:CO ratios (75:25, 50:50 and 25:75) are used and the transition regions for a significant flame acceleration to sonic speed (FA) as well as to a detonation (DDT) are investigated. The conditions for the onset of these transitions are compared with earlier experiments performed in the same facility with H2-air mixtures. The results of this work will help to allow a more realistic estimation of the pressure loads generated by the combustion of H2-CO-air mixtures in obstructed semi-confined geometries.

1.0 INTRODUCTION

The phenomena of effective flame acceleration (FA) and subsequent deflagration-to-detonation transition (DDT) are very important for hydrogen safety considerations, in particular for large semi-confined spaces, such as rooms or tunnels. Such scenarios are also very delicate in nuclear reactor safety, where, e.g. in case of a LOCA MCCI accident, accumulations of released hydrogen at the top of the reactor building might lead to partially confined stratified layers of flammable hydrogen-air mixture [1]. If in a severe nuclear reactor accident also core meltdown occurs, large amounts of hydrogen and carbon monoxide can be formed due to the reaction of molten core and water or concrete. When these gases manage to escape from the safety containment through local leakages that might be caused by the high pressure and temperature loads, flammable H2-CO-air mixtures are generated in adjacent air-filled compartments [2]. The scenario described is not restricted to severe reactor accidents, since similar hazardous situations are also conceivable e.g. in tunnel accidents, where a glowing fire might produce large amounts of CO and large quantities of H2 are released from the damaged tank of a fuel-cell powered vehicle. When ignited, both, H2-air mixtures and H2-CO-air mixtures can lead to high pressure loads during the combustion and might cause further strong structural damage to a facility. For the evaluation of the damage potential that is associated with such scenarios and as a basis for the development of mitigation measures the combustion behavior of such stratified layers in semi-confined geometries has to be investigated. But in contrast to homogeneous H2-air mixtures in closed geometries, rather few experimental data on the combustion behaviour of stratified H2-air-mixtures in semi-confined geometries is currently available, whereas almost no data is available for the combustion behaviour of H2-CO-air mixtures under these conditions.

Fast flames, with velocities in the order of the speed of sound, are identified as a pre-requisite for the transition into a detonation [3], which is the fastest combustion regime of H2-air and H2-CO-air flames and causes the strongest damages to surrounding structures due to the high pressure loads. So, to avoid the onset of a detonation, it is sufficient to prevent a flame from accelerating to sonic speed. The expansion ratio (ratio of specific volume of combustion products over reactants) was identified as a potential for effective flame acceleration (FA) to sonic speed [4]. In earlier experimental investigations on unconfined combustions or combustions with lateral venting a large influence of vents on the flame dynamics was shown [1, 5-7]. The critical conditions for a significant flame acceleration to sound speed

in tubes with lateral venting were expressed using a critical expansion ratio σ*. A linear dependency between this critical expansion ratio and the vent ratio was formulated:

$$\sigma^* = \sigma_0^* \cdot (1+2\cdot\alpha), \quad (1)$$

where σ_0^* - critical expansion ratio for FA in closed tubes; α - vent ratio (vent area/total side walls area). The critical conditions for detonation onset in a tube with lateral venting were found to remain the same as for a closed tube, since the flame velocity reaches sonic speed [8].

In a previous experimental series in a large-scale (9 x 3 x 0.6 m³) horizontal combustion channel with an open ground face an extended criterion for the onset of FA in H2-air mixtures in semi-confined geometries was formulated [1, 7-11]. This criterion was derived by analysing numerous experiments with various layer thicknesses, obstacle configurations and mixture properties. In the resulting graph the expansion ratio of the mixture, representing its thermodynamic properties, is plotted over a dimensionless distance, representing the main geometric properties of the combustion channel. In the graph (left part of Figure 1) open symbols indicate experiments where FA was observed while solid symbols stand for experiments without FA.

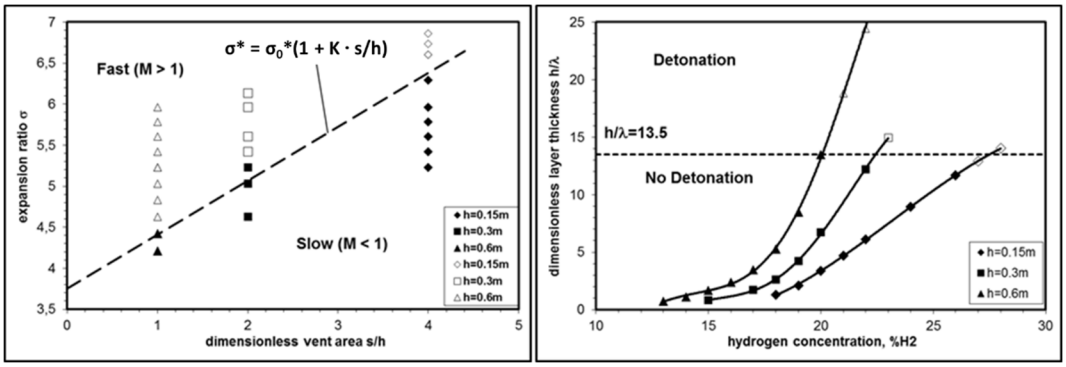

Figure 1. Critical conditions for effective flame acceleration for H2-air mixtures in a semi-confined horizontal channel as function of the expansion ratio [8] (left) and critical conditions for DDT-onset in the same facility as function of the detonation cell size [8] (right).

In the graph the dashed straight line represents the critical expansion ratio σ*, which separates experiments with significant flame acceleration from the experiments without FA. This straight line can be expressed by Equation (2):

$$\sigma^* = \sigma_0^* \, (1 + K \cdot s/h), \quad (2)$$

where σ_0^* is the critical expansion ratio for FA in closed tubes (for hydrogen $\sigma_0^* = 3.75$ [8]); K is a channel-specific constant as a function of integral scale and blockage ratio (for the current channel it was found K = 0.175 [8]); *s* is the obstacle spacing in m and *h* is the layer height in m. The ratio *s*/*h* can be considered as an effective vent ratio for a layer geometry similar to α in Equation (1).

In the same work a criterion for the onset of DDT for H2-air mixtures in semi-confined geometries was proposed using the right graph of Figure 1. In this graph a dimensionless distance is plotted over the hydrogen content of the mixture. Again a dashed line is used to separate experiments with DDT (open symbols in right part of Figure 1) from experiments without DDT (solid symbols in right part of Figure 1). The equation for the horizontal dashed line can be formulated as follows:

$$L \approx 13.5\,\lambda, \quad (3)$$

where *L* is the characteristic length (layer thickness) of the geometry in m; λ is the detonation cell size in m, as a measure of mixture detonability depending on mixture reactivity and gas dynamics of the system. Smaller detonation cell size means higher detonability of the system. According to Equation (3)

a DDT event could occur in experiments where the detonation cell size of the test mixture is at least 13.5 times smaller than the layer thickness of the test mixture. Additionally, also a criterion for the propagation of a detonation through an obstacle was formulated stating that the gap size d of an obstacle has to have at least a height of 3 times the detonation cell size λ to allow a detonation to pass through.

$$d/\lambda = 3, \tag{4}$$

The main objective of the current work is to evaluate the critical conditions for FA and DDT in a semi-confined obstructed horizontal layer of H2-CO-air mixtures and to compare these conditions with results of the previous campaign in the semi-confined horizontal channel with H2-air mixtures.

2.0 EXPERIMENTAL DETAILS

2.1 Test Facility

In the current series of experiments homogeneous H2-CO-air mixtures of various concentrations are ignited in an obstructed horizontal combustion channel with the dimensions 9 m x 3 m x 0.6 m (L x W x H, see Figure 2) in the hydrogen test centre HYKA at KIT. The channel, which has an open ground and rear side, is mounted into a cylindrical safety vessel with a diameter of approx. 3.3 m, a length of 12 m and two hemispherical ends (total volume approx. 98 m³) that is designed for a static pressure of 100 bar. The channel is obstructed by 14 equidistant obstacles (spacing 0.6 m) with a blockage ratio of 50% that are made from 5 horizontal bars with a height of 6 cm and a thickness of 4 cm.

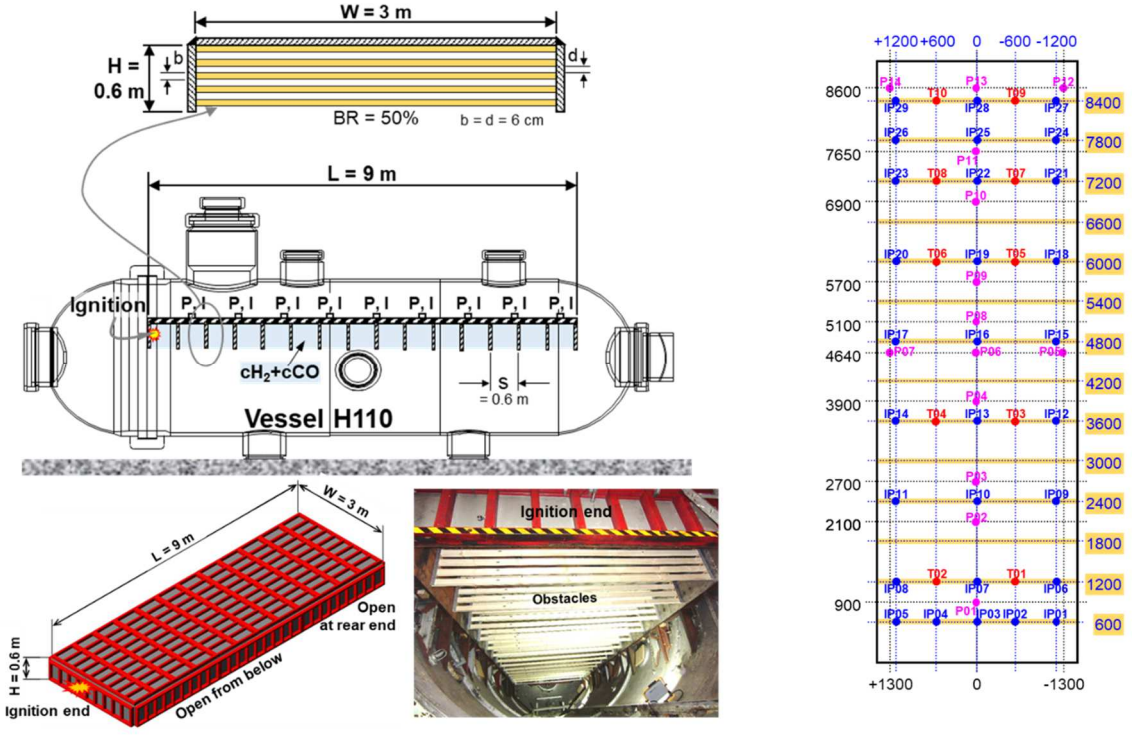

Figure 2. Combustion channel in the safety vessel H110 at KIT-HYKA (left) and sketch of the instrumentation (right).

2.2 Mixture Preparation and Test Procedure

The test mixtures are composed using separate massflow-controllers (Bronkhorst EL-flow series) for the three components H2, CO and air, that were taken from gas bottles (H2 and CO) and the compressed air grid available at KIT (see Figure 3).

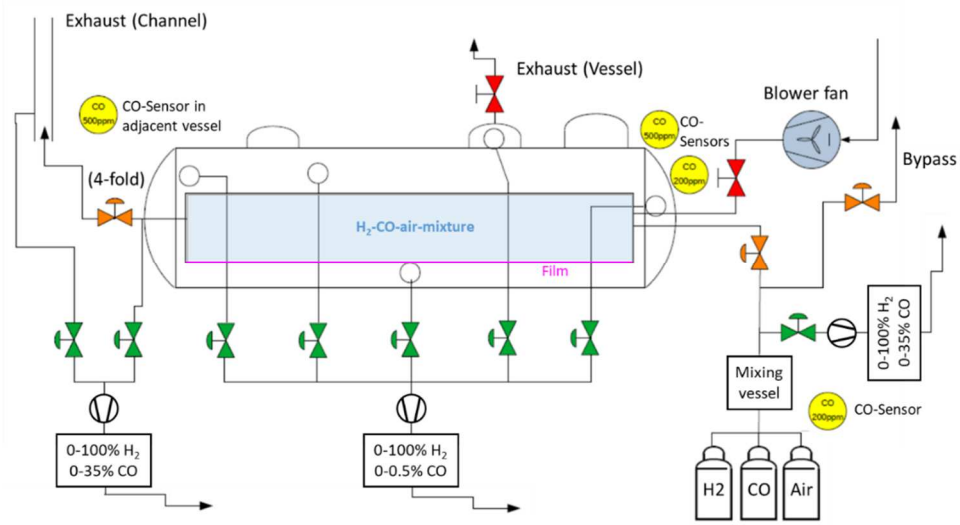

Figure 3. Flow chart of the channel-facility in the safety vessel H110 at KIT-HYKA.

The three flows merge in a small mixing vessel (V = 2.2 dm³), from where they are either conducted through a bypass to the ambience or into the test channel in the safety vessel. Shortly downstream the mixing chamber a small portion of the gas flow is extracted and analysed continuously on the concentrations of the two fuel components in the mixture. At the beginning of an experiment the flow controllers are adjusted to the pre-calculated values to generate the desired mixture composition. During the start-up phase the flow is directed through the bypass line to a chimney until the desired mixture composition is validated by the gas analysis. At this point in time, the flow is switched from the bypass line to the filling line that conducts the mixture into the channel. For the filling procedure the open faces of the combustion channel are closed by a thin plastic film and its sealed volume is then purged through an inlet pipe with many holes at the top of the ignition wall and 4 outlets at the bottom of the rear end. The exhaust atmosphere that contains high CO-concentrations, especially close to the end of the filling procedure, is released to the ambience through a chimney with a height of 10 m above ground level. During the filling procedure the H2-and CO-concentrations in the in- and outlet-flow are monitored continuously, and when the concentrations of in- and outlet are equal the filling procedure is completed. The mixture flow is then conducted through the bypass line again while the flow controllers for the flammable gases are turned off and the pipe system outside the vessel is purged with air.

Although precise instruments are used for the mixture preparation the generation of a ternary mixture in a huge volume of 16.2 m³ is a very delicate issue and despite all care that was taken the concentrations reached at the end of the filling procedure only rarely matched exactly with the desired composition. Furthermore, the result of the settings used shows up only at the end of the procedure, but then changes to the settings are impossible or require further hours of purging to gain the readjusted mixture composition.

2.3 Ignition Procedure and Ignition Device

After the sealed channel is filled with the desired test mixture the experiment is started remotely by an automated routine from a computer in the control stand. When the routine is started the 4 sections of thin plastic film that seal the bottom of the channel are opened simultaneously by x-shaped cuts with 4 sets of 2 heating wires. After the cuts, while the triangular remains of the film are still floating (earlier than centre photo in Figure 4), the ignition source, which comprises of a heating wire in a perforated tube, is turned on. The small flame generated by the glowing wire expands inside the tube but also leaves it through the perforation to produce a broad flame front that covers the complete channel width after a short distance to the ignition wall. Together with the activation of the ignition source a trigger signal is sent to the data acquisition system which then starts to record the signals of the channel instrumentation.

Figure 4. Sealed channel (left), late stage of x-shaped cut of a film segment (center) and ignition device (right).

2.4 Instrumentation

To monitor the combustion processes the channel is equipped with 53 sensors that are mounted in or close to the channel ceiling. In the channel ceiling 14 fast dynamic pressure sensors (PCB, Model M113B23 or similar with ranges from 3.5 bar to 70 bar, P in sketch of Figure 2) are positioned with adapters. At the lower rim of the uppermost obstacle bar in-house fabricated ionization probes and modified thermocouples (IP and T in sketch of Figure 2) for the detection of the flame front are mounted. All sensor signals are recorded simultaneously at a frequency of 100 kHz by a fast data acquisition system (TransCom-RackX by MF Instruments).

2.5 Test Matrix

In the work described it was aimed to provide a data basis on the combustion behaviour of homogeneous H2-CO-air mixtures with different H2:CO-ratios in a semi-confined geometry that can be compared with the results of similar experiments with H2-air mixtures of previous projects in the same facility [1, 8, 9]. Due to the limited resources of the project only 15 experiments were possible, so great care was taken on the choice of the mixture compositions to be investigated. One extra experiment without CO in the test mixture was performed at the beginning of the series to proof the similarity of the current facility with the facility used in the previous projects.

Table 1. Test Matrix of the experiments with homogeneous H2-CO-air mixtures and main mixture properties. Intended concentrations in bold font, real measured concentrations in brackets behind.

Exp.	Fuel [vol%]	cH2 [vol%]	cCO [vol%]	cH2:cCO [-]	σ [-]	λ [mm]
300	***14** (13.6)*	***14** (13.6)*	*0 (0)*	*100:0*	*4.420*	*530.5*
301	**14** (13.9)	**10.5** (10.5)	**3.5** (3.4)	75:25	4.614	440.0
305	**15** (14.9)	**11.25** (11.2)	**3.75** (3.7)	75:25	4.836	285.6
303	**16** (15.8)	**12.0** (11.9)	**4.0** (3.9)	75:25	5.033	195.3
313	**19** (18.9)	**14.25** (14.2)	**4.75** (4.7)	75:25	5.653	55.47
312	**20** (20.0)	**15,0** (14.9)	**5.0** (5.1)	75:25	5.888	3.596
302	**16** (15.9)	**8.0** (8.1)	**8.0** (7.8)	50:50	5.140	158.3
306	**17** (17.0)	**8.5** (8.5)	**8.5** (8.5)	50:50	5.380	97.37
304	**18** (18.0)	**9.0** (9.0)	**9.0** (9.0)	50:50	5.592	63.93
310	**20** (20.1)	**10.0** (10.1)	**10.0** (10.0)	50:50	5.996	33.44
314	**21** (21.0)	**10.5** (10.5)	**10.5** (10.5)	50:50	6.166	25.66
307	**16** (16.0)	**4.0** (4.1)	**12.0** (11.9)	25:75	5.269	151.1
309	**18** (18.1)	**4.5** (4.5)	**13.5** (13.6)	25:75	5.711	68.20
311	**19** (19.2)	**4.75** (4.7)	**14.25** (14.5)	25:75	5.927	49.73
308	**20** (19.8)	**5.0** (5.0)	**15.0** (14.8)	25:75	6.044	41.80
315	**22** (22.1)	**5.5** (5.5)	**16.5** (16.6)	25:75	6.437	26.56

To cover a broad range of mixture compositions it was decided by PS/KIT together with the project partner TUM to concentrate on one layer height of h = 60 cm and two fuel ratios (H2:CO) of 75:25 and 50:50. At a later stage of the project it was agreed that PS/KIT should also try to randomly test the fuel ratio of 25:75, whereas experiments with pure CO as a fuel were ruled out since major difficulties to ignite such mixtures already occurred in the explosion bomb experiments of the first part of the project [13]. Using the results of the previous experiments with H2-air mixtures and the results of the preceding explosion bomb experiments with H2-CO-air mixtures as well as the input from the project partner TUM, where similar tests were made in a smaller facility, the test matrix evolved as the experiments progressed. The mixture compositions of the 15 experiments with three fuel ratios and the one extra-experiment that have been performed in the current work are listed in Table 1. In Table 1 the experiments are not listed chronologically, but according to increasing fuel concentration in groups of increasing CO-fraction in the fuel. As mentioned in chapter 2.2 the preparation of large volumes of a ternary mixture is a delicate issue and thus the concentrations reached at the end of the filling procedure differ slightly from the intended composition in most cases. So the real values measured at the end of the procedure are given in brackets after the intended ones, which are written bold font. To facilitate reading, in the following discussion only the bold numbers will be used when the mixture composition is mentioned.

3.0 RESULTS AND DISCUSSION

3.1 Reference experiment with a H2-air-mixture

The channel facility used in the current work was originally constructed in 2008 for experiments on FA and DDT of H2-air-mixtures in semi-confined geometries. In 2013 it was partly disassembled and rebuilt with a stronger supporting structure to withstand the loads of a second experimental series with a slightly changed focal point. For the current project it was again overhauled and strengthened, and so it was found necessary to perform a reference experiment with a H2-air-mixture to proof that the facility still has the same characteristics as after its first commissioning, after which most of the experiments with H2-air-mixtures in the standard configuration (obstacle configuration, instrumentation) were performed, which will be used for comparison with the current experiments on homogeneous H2-CO-air mixtures.

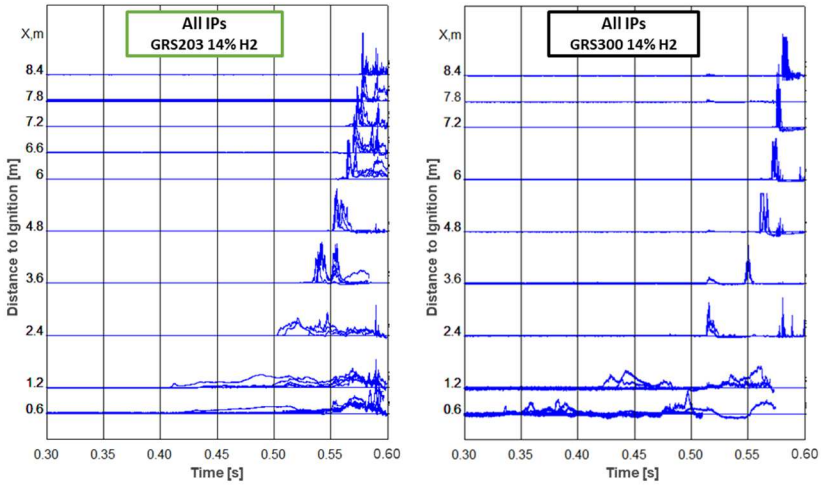

Figure 5. xt-diagrams of the ionization probe signals for the experiments GRS203 (performed in 2013) and the current reference-experiment GRS300.

For the reference experiment a H2-concentration of 14 vol% is chosen that has been the concentration for which FA to sonic speed was just not reached in the first experimental series in 2009. This concentration has the advantage that it allows to check the similarity of the results without taking the risk of strong loads due to a fast deflagration or even detonation. Although a concentration of only 13.6 vol% H2 could be reached in the channel during this first filling procedure with the revised facility the

mixture was ignited and the results are compared with the corresponding experiment of the earlier series. In Figure 5 the signals of the ionization probes for the experiments GRS203 (performed in 2013) and the current reference-experiment GRS300 are compared in the form of xt-diagrams, in which the signals of ionization probes are plotted stacked above each other according to their distance from the ignition source. The comparison of the results of the two experiments performed with similar initial conditions and the same channel set-up shows good agreement for the flame propagation through the channel. For both experiments velocities from 110 m/s to 150 m/s were determined for the last three meters of the channel on the basis of the ionization probe records. For the mean measured pressure in the same region values from 1 bar to 2 bar were determined for both experiments. So with the reference experiment a good similarity of the current facility with the one used in the previous projects was demonstrated, which allows a comparison of the previous and current results.

3.2 Experiments with H2-CO-air-mixtures

To preserve the channel structure it was decided that in the first part of the investigations experiments on the concentration ranges, where a significant flame acceleration to sonic speed (FA) occurs should be investigated, which coincide with the transition from the combustion regime of a slow deflagration to a fast turbulent deflagration. In the second stage experiments on the deflagration-to-detonation transition (DDT) are performed. The results of the experiments with homogeneous H2-CO-air-mixtures in the semi-confined channel facility are summarized in the form of graphs showing the mean velocity determined in the last 3 m of the channel as well as the mean maximum pressures measured by the 5 pressure sensors in the same channel region. In Figure 6 these results and the results of the reference experiment (open black dot) are compared with a series of experiments that were performed in previous projects with H2-air-mixtures (blue diamonds and dashed blue line). In all experiments referred to here the layer height was h = 60 cm.

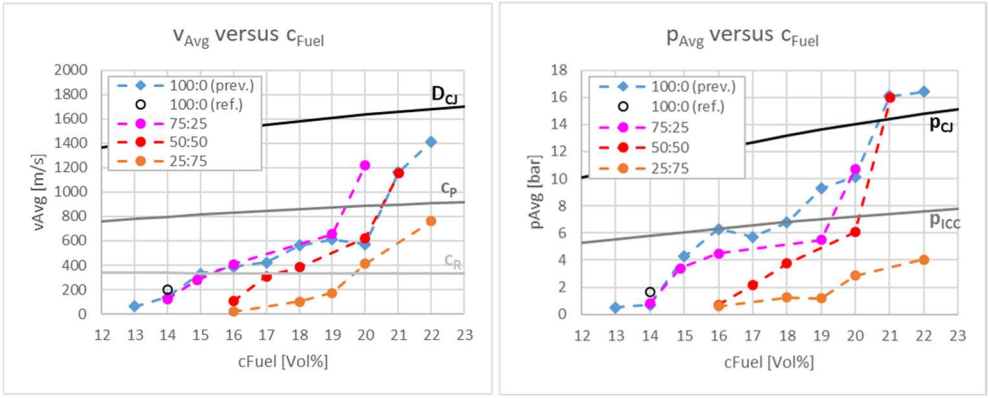

Figure 6. Mean velocity (left, on the basis of IP records) and mean maximum pressures (right) determined in the last 3 m of the channel in the experiments with homogeneous H2-CO-air-mixtures (magenta, red and orange) and H2-air-mixtures (blue and black).

In Figure 6 also the courses of different mixture properties are plotted in dependency of the fuel concentration to facilitate tracking down the transition regions for FA and DDT. These properties are: speed of sound in reactants (c_R) and products (c_P), theoretical Chapman-Jouguet detonation velocity (D_{CJ}), as well as adiabatic isochoric combustion pressure (p_{ICC}) and theoretical Chapman-Jouguet detonation pressure (p_{CJ}). The values for these additional curves were calculated using CANTERA [14] and SDToolbox [15] and are plotted for fuels with a ratio H2:CO of 50:50. The values for other ratios H2:CO do not change more than ±2.5% and do not change the general characterization of flame propagation regime. In the left graph the mean velocities determined for H2-air-mixtures (blue curve), for example, reach the speed of sound in the reactants (c_R in Figure 6) at a hydrogen-concentration of 15 vol%, while the sound speed in the products (c_P in Figure 6) is exceeded at a hydrogen-concentration of 21 vol%. These two concentration values correspond to the concentration values for FA and DDT, respectively, for H2-air-mixtures in semiconfined geometries. The theoretical value for the propagation

speed of a detonation (D_{CJ} in Figure 6) is not reached in the experiments, since the gap in the obstacles is too small to allow the detonation to pass through, as the detonation propagation criterion described by Equation (4) demonstrates. According to Equation (4) a mixture with a detonation cell size of $\lambda \leq 2$ cm is required to allow the detonation to propagate through the obstacles. But the mixtures with H2-concentations of 21 vol% and 22 vol% used in the experiments have detonation cell sizes of 3.2 cm and 2.5 cm, respectively, and thus the detonation quenches in the obstacle and has to be reinitiated shortly behind it. Due to this quenching a velocity deficit occurs in all experiments described here. For the pressure values plotted in the right graph of Figure 6 pressure values in the range of the theoretical Chapman-Jouguet detonation pressures velocities (p_{CJ} in Figure 6) are measured, since all pressure sensors are located in positions in between two obstacles, where the detonation can propagate undisturbed. But in H2-CO-air-mixtures the adiabatic isochoric combustion pressure (p_{ICC} in Figure 6) is only approached but not exceeded when the onset of FA is detected in the velocity graphs.

The data point for the reference experiment of the current project with a H2-concentation of 14 vol% (open black circle in Figure 6) fits quite well with the corresponding experiment of the earlier series with H2-air-mixtures (blue diamond in Figure 6), although it lies slightly higher in both graphs.

The experiments with hydrogen rich H2-CO-air-mixtures with a fuel ratio H2:CO of 75:25 (magenta-line in Figure 6) also show quite good agreement with the course of the experiments with H2-air-mixtures. Significant flame acceleration to sonic speed is firstly observed at a fuel-concentration of approx. 15 vol% to 16 vol%, which is almost the same value as for the test mixture without CO. For the mixtures with this fuel ratio furthermore DDT is observed even at a slightly lower fuel-concentration of approx. 20 vol% compared to the mixtures without CO (21 vol% H2). For the H2-CO-air-mixtures with a fuel ratio H2:CO of 50:50 (red line in Figure 6) the transition to a flame with sonic velocity was observed for a fuel concentration of approx. 17 vol% to 18 vol%, which is about 2 vol% higher than for H2-air-mixtures, while DDT was observed for a mixture with a fuel-concentration of 21 vol%, which is the same fuel-concentration as for pure H2-air-mixtures. For the randomly tested carbon-monoxide rich H2-CO-air-mixtures with a fuel ratio H2:CO of 25:75 (orange line in Figure 6) only the transition to a fast sonic flame could be determined, which was observed for a fuel concentration of approx. 20 vol%, which is about 5 vol% higher than for H2-air-mixtures. Thus, the higher CO content in a fuel suppresses the flame acceleration and detonation transition.

3.3 Flame acceleration (FA) to sonic speed in H2-CO-air-mixtures

The transition from a slow to a fast turbulent deflagration can be identified by a strong pressure increase to loads in the region of p_{ICC} and flame velocities in the range of c_R in the experimental records. In the previous work with the same facility a critical expansion ratio σ^* for H2-air mixtures in semi-confined geometries was introduced, which can be calculated using Equation (2). In (2) the geometrical properties of the facility are summarized as ratio of obstacle spacing and layer height (s/h) and a channel-specific constant K. It was found that for H2-air mixtures with an expansion ratio σ larger than the critical expansion ratio σ^* flame acceleration to sonic burning velocity becomes possible. In the current work with H2-CO-air mixtures a similar approach was chosen and so apart from the mixture composition also the expansion ratio σ of every test mixture, calculated with CANTERA [14] and SDToolbox [15], is given in Table 1.

In Figure 7 the expansion ratios of the H2-air mixtures and H2-CO-air mixtures in semi-confined geometries already shown in Figure 6 are plotted over the CO-fraction in the fuel. In the graph open symbols correspond to experiments in which flame acceleration to sonic speed was observed, while closed symbols correspond to experiments without FA. The dashed yellow line in Figure 7 represents the σ-criterion for FA in semi-confined geometries that was developed in the previous projects (Eq. 2) [1, 7-12] in an extension for two fuel components that are weighted according to their molar fraction:

$$\sigma^* = [H_2] \cdot \sigma_0^*(H_2) \cdot \left(1 + K\frac{s}{h}\right) + [CO] \cdot \sigma_0^*(CO) \cdot \left(1 + K\frac{s}{h}\right) \quad (5)$$

The fuel composition is considered by the molar fractions [H2] and [CO] of the fuel components in the total fuel concentration, and σ_0*(H2) as well as σ_0*(CO), which are the critical expansion ratios for binary mixtures of the components H2 and CO in air for closed geometries.

To gain a value for the unknown critical expansion ratio for CO-air mixtures in closed geometries σ_0*(CO), earlier experiments with H2-CO-air-mixtures in a closed tube with rectangular cross-section were used [16]. In the experiments the combustion behaviour of homogeneous H2-CO-air-mixtures with H2:CO-ratios of 100:0, 75:25 and 50:50 was investigated in a limited fuel-concentration range from 9 – 15 vol%. The critical expansion ratios σ_0* for these compositions were determined using Equation (6),

$$v_{max}/c_P \approx 0.8 \quad (6)$$

with v_{max} being the maximum flame speed and c_P representing the speed of sound in the burned gas. Extrapolating these values to the H2:CO-ratio of 0:100 yields a value of σ_0*(CO) = 4.65 for the critical expansion ratio of CO-air-mixtures in closed geometries.

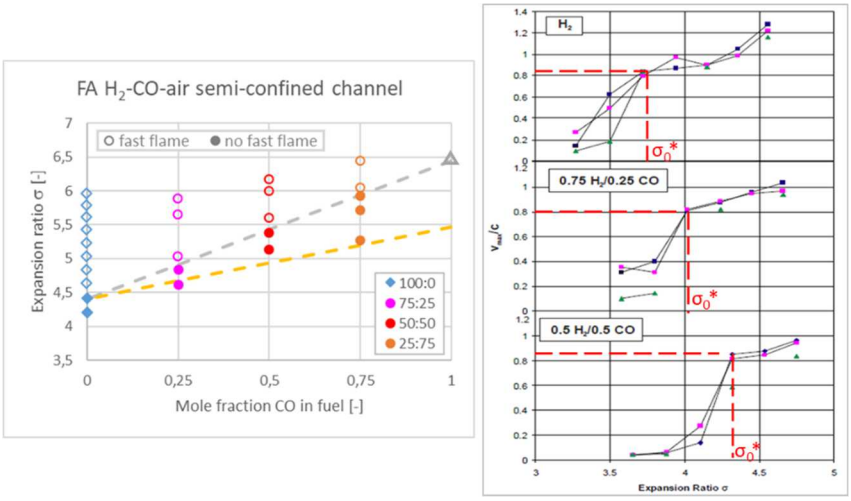

Figure 7. Left: Expansion ratios σ of the tested H2-air-mixtures and H2-CO-air-mixtures over fuel composition and extended criteria for FA in these mixtures (dashed lines), Right: results of earlier experiments with H2-CO-air-mixtures in a closed tube with rectangular cross-section [16].

Similar to the left graph of Figure 1, the dashed yellow line in Figure 7, representing Equation (5), separates regions in the graph where flame acceleration was observed (upper left part of the graph) or not (lower right part). Figure 7 demonstrates, that the extended criterion of Equation (5) is still accurate for mixtures without CO, but it increasingly overestimates the potential for flame acceleration to sonic speed with increasing CO-fraction in the fuel. The discrepancy grows almost linearly with increasing CO-fraction, so it seems that a pre-factor *a*, containing fuel specific properties, is missing. Since Equation (5) is accurate for H2-air-mixtures, the pre-factor for hydrogen can be set to *a*(H2) = 1. To determine the pre-factor for CO a graphical solution is chosen, in which the value for the critical expansion ratio of CO-air-mixtures in the semi-confined channel is set to be σ*(CO) ≈ 6.45 in the graph (grey triangle in Figure 7). With these settings a pre-factor of *a*(CO) = 1.18 is determined, which generates values for σ* that correspond with the dashed grey line in Figure 7, which now properly separates experiments with FA from experiments without FA. The equation for the grey dashed line in Figure 7, which corresponds to the modified FA-criterion for H2-CO-air-mixtures in semi-confined geometries can be written in the form of Equation (7):

$$\sigma^* = a(H2) \cdot [H_2] \cdot \sigma_0^*(H_2) \cdot \left(1 + K\frac{s}{h}\right) + a(CO) \cdot [CO] \cdot \sigma_0^*(CO) \cdot \left(1 + K\frac{s}{h}\right) \quad (7)$$

In this equation the pre-factors *a*(H2) and *a*(CO) can be interpreted as a measure for the "accelerability", or tendency of a flame to accelerate in a binary mixture of the respective fuel component in air with respect to the behaviour of H2-air mixtures. It is conceivable that *a* incorporates all properties of the compound that may influence the tendency for FA, but are not covered by the expansion ratio σ, as for example kinetics, inhibitions or influences of reaction pathways. Pre-factors $a > 1$ indicate that the tendency to accelerate is weaker than for H2, since such values lead to higher critical expansion ratios σ* for FA, while values $a < 1$ indicate that the tendency to accelerate is more pronounced than for H2.

3.4 Deflagration-to-Detonation Transition (DDT) in H2-CO-air-mixtures

The second step in the experimental velocity and pressure courses of Figure 6 correspond to the transition from a fast turbulent deflagration to a detonation, but this transition region was only identified for the H2:CO fuel compositions of 75:25 and 50:50. Due to time pressure, the limited budget of the project, the vulnerability of the channel in the detonation experiments and the out-of-scope position of the H2:CO fuel composition of 25:75 for nuclear safety it was decided to stop the investigations on this fuel composition after no detonation was observed for a fuel content of 22 vol% in air, since this experiment had proven that DDT is shifted to higher concentrations compared to the other mixture compositions. As shown in Figure 1 and Equation (3), also a criterion for the onset of DDT for H2-air-mixtures in semi-confined geometries was formulated in the previous projects that takes into account the layer geometry (layer height h) and the reactivity of the mixture (detonation-cell-size λ). To check the applicability of this criterion on the H2-CO-air mixtures investigated in the current project a graph similar to the left graph of Figure 7 is shown in Figure 8.

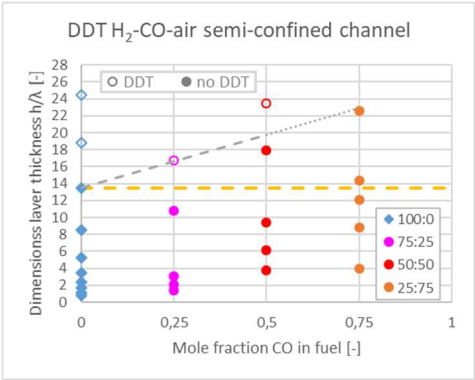

Figure 8. Dimensionless layer thicknesses for H2-air-mixtures of pervious projects (blue) and current H2-CO-air-mixtures (magenta, red and orange) over fuel composition and extended criteria for DDT in these mixtures (dashed yellow and grey lines).

In this graph the DDT-criterion for H2-air-mixtures in semi-confined geometries of Equation (3) is plotted as dashed yellow line. Similarly to Figure 7, the criterion works quite well for H2-air-mixtures without CO, but for H2-CO-air mixtures the possibility for DDT is increasingly overestimated with increasing CO-fraction in the fuel. Unfortunately, no data on detonation-cell-sizes for CO-rich H2-CO-air mixtures is available, so an empirical approach was chosen to fit at least the data available for the fuel compositions of 100:0, 75:25 and 50:50 and the indications from the experiments with a H2:CO fuel composition of 25:75 (no DDT observed for mixtures with up to 22 Vol% of fuel). So in Equation (8) a separation due to the different fuels and a weighting with respect to the molar fractions [H2] and [CO] of the combustible components in the fuel is proposed.

$$\frac{h}{\lambda} = [H_2] \cdot \frac{h}{\lambda(H_2)} + [CO] \cdot \frac{h}{\lambda(CO)} \tag{8}$$

The value of h/λ(H2) = 13.5 for a mixture without CO is given by the criterion of Equation (3), so the slope of the dashed grey line that should separate regions with DDT from regions without DDT in Figure 8 can be adjusted by the value for h/λ(CO). When a value of h/λ(CO) = 26 is applied the line

separates all data points of the current series correctly, but, of course, further data points, especially for CO-rich mixtures, are needed to proof the reliability of Equation (8) over the complete mixing range. This is the reason why the dashed grey line is drawn dotted for CO-molar fractions above 0.5, and is even omitted for mixtures with more than 75% of CO in the fuel.

3.5 Evaluation of the results

In Figure 6 the results of the current experiments with H2-CO-air-mixtures are plotted against the total fuel concentration, which corresponds to a scenario where H2 is replaced by CO in the mixture. In this representation the measurement points for the mixtures with only 25% CO in the fuel (magenta symbols), agree very well with the corresponding values for H2-air-mixtures (blue symbols) of the earlier series. Only in the test with 20% fuel in the mixture (H2:CO = 75:25) significantly higher velocities are determined. In contrast, the tests with higher CO fractions of 50% and 75% CO in the fuel gas (red and orange symbols, respectively) show significant deviations mainly for the transitions from slow to fast deflagration (approx. 15 vol% for H2:CO = 75:25, approx. 17 vol% for 50:50 and approx. 20 vol% for 25:75), but also, albeit to a much lesser extent, the transitions to a detonation. DDT for mixtures with 75% H2 in the fuel gas was determined at approx. 20 vol% fuel, which is in line with the value found in the past for H2-air-mixtures (approx. 21 vol%). For mixtures with 50% CO in the fuel gas fraction, DDT has been determined at 21 vol% fuel, which also agrees well with the value for H2-air-mixtures. For the mixture ignited in the last experiment in this series with a fuel content of 22 vol% with a fraction of 75% CO in the fuel gas, on the other hand, no detonation could be observed. However, in view of the great damage this and the previous experiments had caused to the channel, it was decided to perform no further experiments with even higher fuel contents with this fuel gas composition, as the result obtained already confirmed the observed trend.

When the same velocities and pressures are plotted against the H2 content in the mixture, the representation corresponds to scenarios, where CO is added to an existing H2-air-mixture, see Figure 9. Due to the addition of CO to the mixture, fast turbulent deflagrations are already possible at H2 concentrations of about 5 vol%, 9 vol% and 12 vol% when certain amounts of CO (here: 3xcH2, 1xcH2 and 0.33xcH2, respectively) are added. However, the influence of the CO addition is even more obvious with regard to the onset of a detonation. While a concentration of 21 vol% H2 in the mixture is required for detonation in H2-air-mixtures, detonations can already be observed in H2-CO-air-mixtures at H2 concentrations of 15 vol% and 11 vol%, respectively, if the fuel content of the mixture is increased to approx. 21 vol% by the addition of CO.

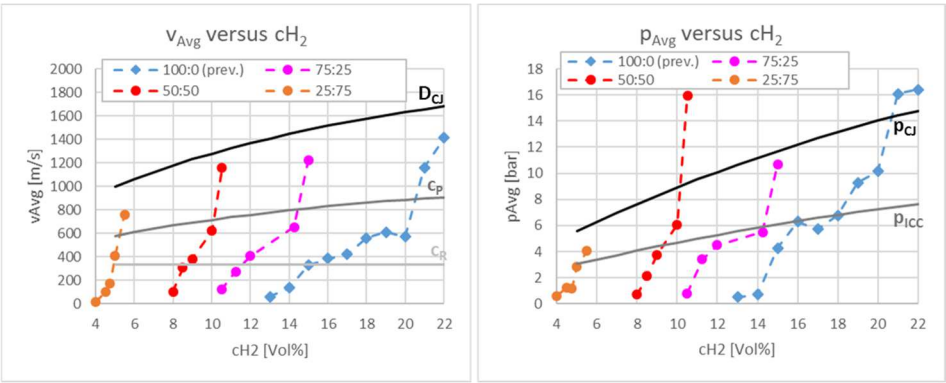

Figure 9. Mean velocity (left, on the basis of IP records) and mean maximum pressures (right) determined in the last 3 m of the channel in the experiments with homogeneous H2-CO-air-mixtures (magenta, red and orange) and H2-air-mixtures (blue and black) versus H2-concentration.

4.0 SUMMARY AND CONCLUSION

To evaluate the influence of CO on the critical conditions for an effective flame acceleration (FA) to speed of sound and the deflagration-to-detonation transition (DDT) in semi-confined obstructed

horizontal layers of H2-air mixtures numerous combustion experiments have been performed in a horizontal channel (9 x 3 x 0.6 m³) at KIT. In the tests, slow subsonic and fast sonic deflagrations as well as detonations were observed and the conditions for FA and DDT were determined and compared with the results of previous campaigns with H2-air mixture layers in the same facility.

Based on experiments with H2-air mixtures in semi-confined horizontal layers Equation (2) had been formulated as a criterion for the onset of FA. In this equation the expansion ratio σ is used as measure for the mixture reactivity. In the current work the criterion was extended to H2-CO-air mixtures, by taking the different combustion properties of H2 and CO into account by weighing their influence on the critical expansion ratio for semiconfined geometries σ* according to their molar fraction in the fuel. Furthermore, a pre-factor *a* was introduced for every fuel component that incorporates all properties relevant for FA that are not covered by the expansion ratio (e.g. kinetics, inhibitions, reaction pathways) with respect to the FA behaviour of H2 (a(H2) = 1). In its modified form and with a pre-factor of a(CO = 1.18) the criterion could be successfully applied to all experiments of the current series.

The criterion for DDT in semi-confined horizontal H2-air layers formulated previously in Equation (3), in which the detonation cell size λ represents the mixture reactivity, can also be adapted to H2-CO-air mixtures. In this case the unchanged criterion yields a very conservative evaluation of CO-containing mixtures, since the possibility for DDT is more and more overestimated with increasing CO-fraction in the fuel. An approach for a more accurate evaluation concerning DDT in semi-confined H2-CO-air mixtures was proposed, but this approach still lacks data on the detonation behaviour of CO-rich mixtures with more than 50% of CO in the fuel for justification over the complete mixing range.

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