

Turbulent explosions in H₂ enriched CO and CH₄ mixtures

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Abstract

The use of hydrogen enriched fuel streams has come to the fore due to the use of syngas and/or biogas related feedstocks in gas engine or gas turbine based power generation applications. The associated safety implications are, however, not well established. The current work presents a systematic study of over-pressures generated in an obstructed explosion tube. The conditions were chosen to generate strongly turbulent explosions with resulting flame speeds in the range 200 to 300 m/s. The use of hydrogen enrichment also facilitates the use of fuel lean to ultra-fuel lean blends due to overall improved combustion characteristics. Accordingly, the current focus is on such mixtures with CH₄/H₂ and CO/H₂ systems investigated for hydrogen enrichment levels between 0 and 100% and for stoichiometries of 0.40, 0.60 and 0.80. The results highlight the differences in behaviour between the two blending components, with CO mixtures providing substantially higher over-pressures than the corresponding CH₄ blend. The results suggest that methane has a mitigating effect up to comparatively high hydrogen blending fractions and that synergistic effects between fuel components need to be taken into account.

1 Introduction

The over-pressure in an explosion is determined by the flame propagation speed which is primarily governed by the flow condition as well as the mixture reactivity. Obstacles in the flame propagation path can accelerate the flame speed by introducing turbulence in the recirculation zone. The turbulence will increase the rate of heat release due to interface wrinkling resulting in an increased flame front area. The result is an increase in the the flow velocity, in turn causing stronger turbulence which, further, increases the rate of heat release. This positive flame acceleration feedback mechanism is well established. For example, Moen et al. [1, 2] obtained flame speeds up to 130 m/s in stoichiometric methane-air mixtures – around ~24 times the flame speed observed in the absence of obstacles. Repeated obstacles can accelerate the flame speed continuously and trigger a transition from deflagration to detonation [3, 4]. The controlling factor of this mechanism was attributed to the blockage ratio with the flame speed relatively insensitive to the manner by which the turbulence is generated [1]. The blockage ratio influences the maximum over-pressure while the rate of increase depends on the obstruction geometry. The wall/plate type obstructions lead to the highest over-pressures and the cylindrical obstruction yields the lowest over-pressure according to Ibrahim and Masri [5]. The influence of the obstacle separation distance was studied by Abdulmajid et al. [6]. The interest in hydrogen enriched mixtures has resulted in comparatively recent studies by Lowesmith et al. [7] and Bauwens et al. [8]. The current data sets feature a comparatively simple two-obstacle layout aimed at facilitating further computational investigations in order to enable an assessment of the ability of computational models to reproduce the systematic differences between the behaviour of methane and carbon monoxide blends with hydrogen. Furthermore, the geometry is a development of the single obstacle configuration used by Lindstedt and Sakthitharan [9] to obtain a directly related quantitative description of the interaction of the flame with obstacles by measuring the time-resolved velocities in a two-dimensional plane along the vertical tube axis. Reynolds

stress components were obtained. Despite the comparatively large number of investigations, comprehensive data sets that explore the current systems are absent. However, methane–air mixture have been frequently used [1, 2, 6] along with liquefied petroleum and liquefied natural gas related fuel components [3–5]. In this work, the over–pressures resulting from obstruction-enhanced explosions have been experimentally studied with extended range of gases. Binary fuels of lean CO/H₂/air and CH₄/H₂/air mixtures over a wide range of hydrogen content were used and a database was obtained. The current comprehensive test matrix is shown in Table 1.

Table 1: Test matrix

Φ	$\alpha(H_2, CO)$	$\alpha(H_2, CH_4)$
$\Phi=0.80$	0.1 0.2 0.3 0.4 0.5	0.0 0.2 0.4 0.5 0.6 0.7 0.8
$\Phi=0.60$	0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0	0.2 0.4 0.5 0.6 0.7 0.8 0.9 1.0
$\Phi=0.40$	0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0	0.7 0.8 0.9 1.0

The hydrogen content is defined in the customary manner $\alpha = \frac{C_H}{C_H + C_F}$, where C_H and C_F represent the mole fractions of H₂ and CO/CH₄, respectively. The conventional composition parameter imposes of disproportionately larger measure of the hydrogen concentration on the flame response as pointed out by Wu et al. [10]. It is because, compared to hydrocarbons, the oxidation of per mole of H₂ needs less oxygen, releases less heat and heats up less nitrogen. Yu et al. [11] proposed another parameter to represent the relative amount of hydrogen addition, shown in Eq. 1.

$$R_H = \frac{C_H + C_H / (C_H / C_A)_{stoic}}{C_F + [C_A - C_H / (C_H / C_A)_{stoic}]} \quad (1)$$

This formulation has been shown to be able to facilitate data reduction and correlation [10–12]. However, it was only tested for mixtures with low H₂ content. A modified form was found useful for the current extended conditions with much higher H₂ content with the parameter β defined as below,

$$\beta = \left[\frac{C_H}{(C_H / C_A)_{st}} \right] / \left[\frac{C_H}{(C_H / C_A)_{st}} + \frac{C_F}{(C_F / C_A)_{st}} \right] \quad (2)$$

where C_H is the H₂ concentration, C_F the fuel concentration (CO or CH₄), $(C_F / C_A)_{st}$ is the concentration at stoichiometric and of pure fuel components (i.e. for CH₄: $(C_F / C_A)_{st} = 0.105$, for CO: $(C_F / C_A)_{st} = 0.420$ and for H₂: $(C_H / C_A)_{st} = 0.420$).

2 Experimental Setup

The experiments were carried out in a flame driven shock tube (4.420 m × 0.034 m × 0.072 m) as schematically shown in Fig. 1. Two solid obstacles with a height of 0.036 m, offering a blockage ratio of 50%, were installed at distances of 0.080 m and 0.360 m from the ignition end and located on the top and bottom in a staggered arrangement in order to enhance the flame propagation. The large ratio of shock tube length to hydraulic diameter (~ 96) allows the investigation of the flame obstacle interaction without interference from acoustic waves reflected off the closed end. Nine ionisation probes were mounted along the shock tube and served as flame arrival time detectors. The ionisation probes were made of coaxial stainless steel cable with a resistance $> 10 \text{ M}\Omega$ between the core and the shield and an outer diameter of 1.5 mm. The pressure was measured using four piezo–electric pressure transducers (1 × PCB 113A21 and 3 × PCB 113B21; PCB Piezotronics, Inc.). A 12-bit data acquisition card (PCI-6115; National Instruments) with a recording rate of 1 MHz was used for both ionisation probes and pressure transducers and all the signals were recorded for further analysis. The positions of the ionisation probes and pressure transducers are shown in Table 2.

Table 2: Port location (X) for installed pressure transducers (P) or ionisation probes (I).

Port	PN1	PN2	PN3	PN4	PN5	PN6	PN7	PN8	PN9
X [m]	0.845	1.075	1.305	1.535	1.765	2.000	2.225	3.145	3.450
Type	P+I	I	P+I	I	I	I	P+I	P+I	I

Before each experiment, the shock tube was flushed with air for about one minute and then evacuated to a pressure below 0.5 kPa. The fuel components and air were introduced and measured using the partial pressure method to prepare the target mixtures. The pressure was monitored by a static pressure transducer (UNIK 5000; GE Measurement & Control). Subsequently, the gas mixture was circulated for five minutes, corresponding to 20 shock-tube-volumes, in order to achieve homogeneity. Prior to ignition, the mixture was left to settle for two minutes to achieve quiescent conditions. The whole process was controlled by a purpose written LabView program and all data acquisition devices were triggered using a TTL pulse to ensure synchronisation. For each data point, three separate experiments were carried out to provide an estimate of the experimental reproducibility.

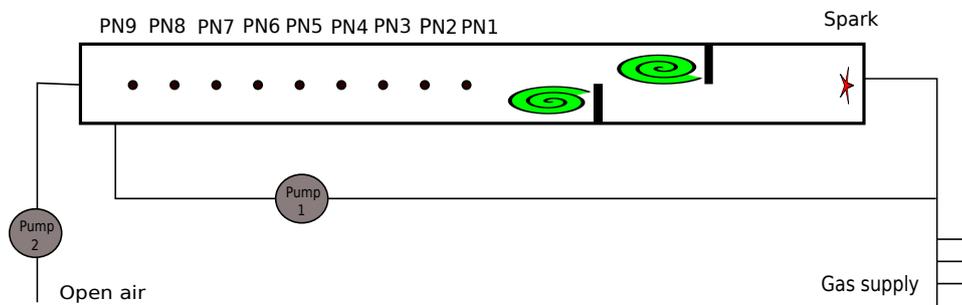


Figure 1: Schematic of the experiment setup; PN – Port Number

Examples of flame arrival times as detected by the ionisation probes is shown in Fig. 2(a) together with the calculated flame speeds for a CO/H₂/air mixtures at $\Phi = 0.80$. The dashed line separating the graph into Zones A and B, represents the estimated location where the pressure wave back reflection interacts with the flame front. The values in Zone B are influenced by reflected wave interactions and are hence not used. The resulting flame speeds are shown in Fig. 2(b).

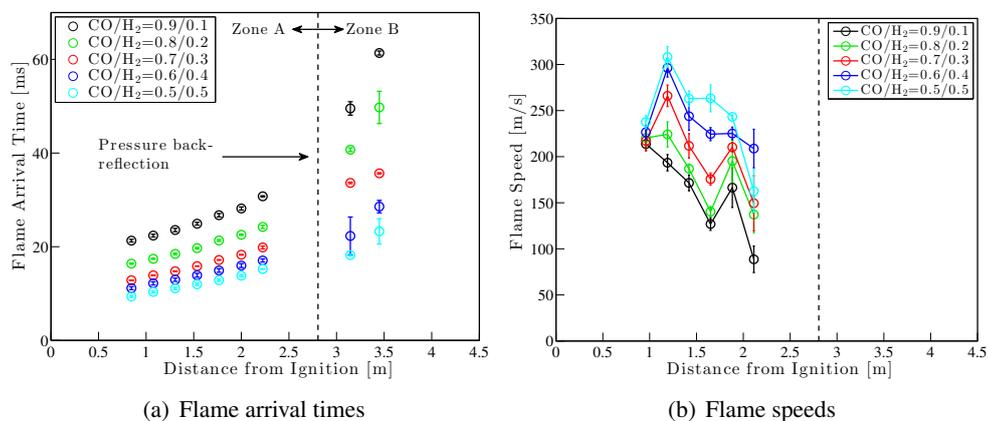


Figure 2: Flame arrival times and flame speeds of CO/H₂/air mixtures at a stoichiometry $\Phi = 0.80$

The flame speed increases with H₂ percentage due to its higher reactivity. Yet, it is interesting to note

that the influence is not prominent at the first measurement point while most significant at the second, i.e. 1.190 m from the ignition end. The same trend was found for $\Phi = 0.60$ but featuring longer flame arrival times and lower flame speeds. For mixtures with $\Phi = 0.40$, the ionisation probes did not trigger reliably due to low ion concentrations.

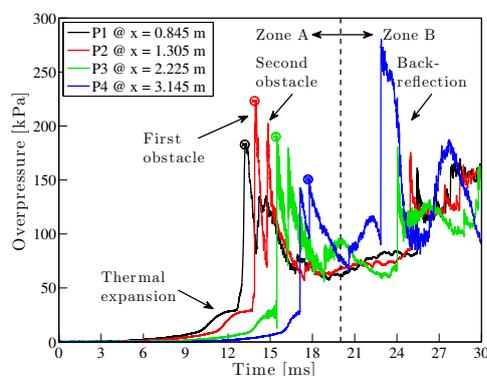


Figure 3: Explosion over–pressure traces in CO/H₂/air mixture ($\Phi = 0.80$ CO/H₂ = 0.7/0.3)

3 Results and Discussion

The initial laminar flame kernel is caused by an electric spark with an energy level of approximately 5 mJ [3,4,9,13] and the subsequent flame front propagates semi-spherically until it reaches the wall of the shock tube. Subsequently, the flame front propagates forward consistent with the velocity profile [9].

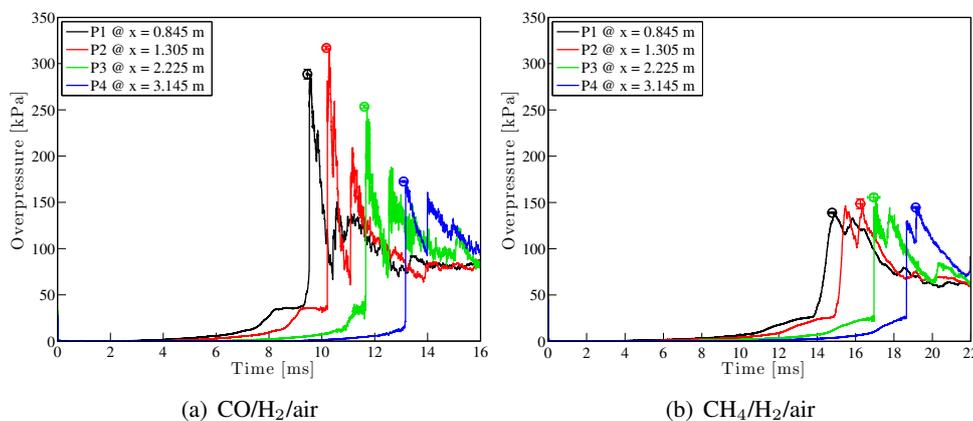


Figure 4: Explosion over–pressures at a stoichiometric $\Phi = 0.80$ and H₂ content of 50%

Hampp et al. [14] visualised the initial flame propagation via high speed chemiluminescence and high speed PIV in the same geometry with slightly different obstructions. The thermal expansion of the hot combustion products behind the flame front increases the flame speed and also causes a slight pressure rise. The thermal expansion causes a combustion driven flow in front of the flame front, leading to strong turbulence generation in the shear layer and recirculation zone behind the obstacle. The result is two strong consecutive turbulence driven explosions (behind each obstacle) leading to a sharp pressure rises. The explosion over–pressure was measured at four different locations along the shock tube.

The pressure development is exemplified in Fig. 3 using CO/H₂/air mixtures at $\Phi = 0.80$ and a H₂ content of 30%. Data for all four pressure transducers are shown. The two peak pressures of each transducer

corresponds to the explosions behind the two obstacles as marked in Fig. 3. The initial pressure rise of approximately 25 kPa is caused by the thermal expansion associated with the initial flame propagation. The signals in Zone B, which are somewhat chaotic, are the result of the pressure back-reflection from the closed end of the shock tube.

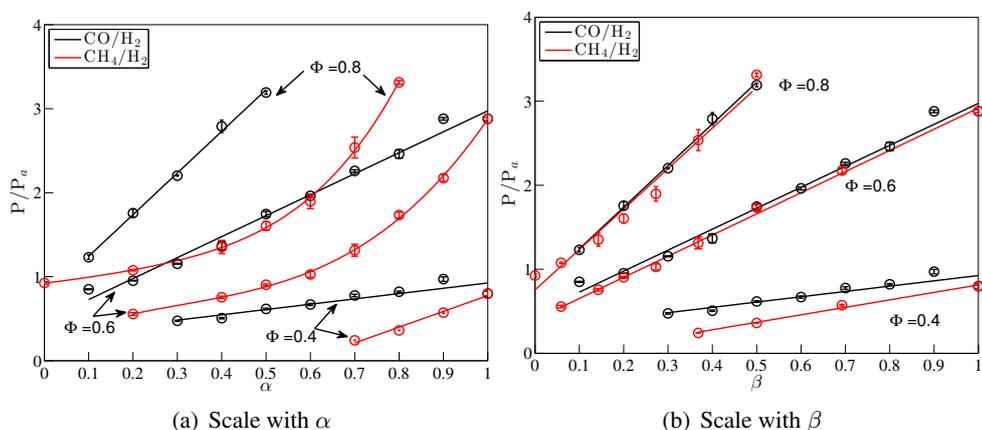


Figure 5: Influence of H₂ content on the peak pressures at a distance of 1.305 m from the ignition end. Data obtained by the second pressure transducer. The symbols denote experimental data and the lines simple fits.

The pressure rise in CO/H₂/air and CH₄/H₂/air mixtures with $\Phi = 0.80$ and H₂ fraction of 50% is shown in Fig. 4. The peak pressures for CO/H₂ mixtures are considerably higher than for the CH₄/H₂ mixtures with shorter peak pressure arrival times. This shows that CO is more reactive than CH₄ and also suggests that CH₄ has a stronger inhibiting effect than CO on the combustion chemistry of H₂. The pressure rise caused by the initial flame propagation for the CO/H₂ mixtures is notably higher than that for CH₄/H₂ mixtures, despite their similar thermal expansion factor ($\tau = 6.54$ for CO/H₂/air, $\tau = 6.45$ for CH₄/H₂/air). Consequently, the deviation can be attributed to the faster undisturbed flame acceleration due to chemical effects. The small error bars (based on a minimum of three experiments at each data point) further show the high repeatability obtained in the current configuration.

A direct comparison of the influence of hydrogen content and equivalence ratio on the peak over-pressure in CO/H₂/air and CH₄/H₂/air mixtures is shown in Fig. 5. The success of the β scaling in explaining the behaviour of the two mixtures is apparent and of direct relevance to risk assessment of hydrogen enriched fuels. For all mixtures, the over-pressure increases linearly with β for all stoichiometries and the increase is faster at higher equivalence ratio. The significant difference in the actual behaviour for the over-pressure, see Table 3, is possibly due to a catalytic effect of H₂ on the oxidation of CO and/or that CH₄ has an inhibiting effect on hydrogen at comparatively low concentration levels. It is suggested that, in the context of risk assessments, not only the hydrogen content and the mixture stoichiometry is of relevance, but also the chemical interactions between the different fuel components.

4 Conclusions

In order to assess the risk associated with hydrogen enrichments found in the use syngas, biogas and natural gas systems, a series of experiments were performed using fuel lean premixed CO/H₂/air and CH₄/H₂/air mixtures at various compositions. The equivalence ratio was varied from 0.80, 0.60 to 0.40. The hydrogen content was varied from close to the flammable limit to mixtures producing strong turbulent explosions. The experiments were carried out in a shock tube configuration capable of high reproducibility with explosion over-pressures and flame arrival times measured. The following conclusions can be drawn:

- Both hydrogen content and equivalence ratio have a significant influence on the explosion severity in CO/H₂/air and CH₄/H₂/air mixtures. The explosion over-pressure increases with hydrogen content and equivalence ratio.
- The over-pressure increases linearly with β for all mixtures but with different slopes.
- At the same equivalence ratio and hydrogen content, CO/H₂/air mixtures give a higher explosion over-pressure than CH₄/H₂/air mixtures, illustrating a stronger inhibiting effects of CH₄ than CO on the combustion of H₂.
- The flame arrival time decreases with an increase in both equivalence ratio and hydrogen content and the impact on flame speed has been quantified.

The current work suggest that ternary fuels blends (e.g. CO/CH₄/H₂) should also be investigated due to the apparent importance of interactions between the chemistries of different fuels.

Table 3: Peak explosion over-pressures

CO/H ₂ /Air	$\Phi=0.80$	H ₂ (%)	10	20	30	40	50	-	-	-	-
		P (kPa)	125	178	223	283	323	-	-	-	-
	$\Phi=0.60$	H ₂ (%)	10	20	30	40	50	60	70	80	90
		P (kPa)	86	96	117	139	177	199	229	249	292
	$\Phi=0.40$	H ₂ (%)	-	-	30	40	50	60	70	80	90
		P (kPa)	-	-	48	51	62	68	79	83	98
CH ₄ /H ₂ /Air	$\Phi=0.80$	H ₂ (%)	0	20	40	50	60	70	80	-	-
		P (kPa)	94	109	137	163	192	257	336	-	-
	$\Phi=0.60$	H ₂ (%)	-	20	40	50	60	70	80	90	100
		P (kPa)	-	56	77	92	104	133	176	220	292
	$\Phi=0.40$	H ₂ (%)	-	-	-	-	-	70	80	90	100
		P (kPa)	-	-	-	-	-	25	37	58	81

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