TURBULENT FLAME PROPAGATION IN GAS MIXTURES

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Both offshore and onshore process streams involve in many occasions mixtures of gases. An important example is natural gas which is a mixture of methane, ethane and some higher hydrocarbon gases. It was therefore decided to start investigations into the turbulent explosion properties of mixtures of gases using wedge-shaped vessels with obstacles. The investigation showed that the turbulent burning rate of mixtures of two gases lies in between the turbulent burning rate of the pure gases. The turbulent burning rate of natural gas can be represented satisfactorily by the turbulent burning rate of mixtures of methane and ethane or methane and propane.

Key words: gas explosion, turbulence, scaling, burning velocity

INTRODUCTION

Gas explosions have shown to have a very high damage potential. A review of the hundred largest losses in the hydrocarbon process industry, from 1957 to 1986 was performed by Garrison (1). He found that 42% of these accidents were caused by vapour cloud explosions. In his classification vapour cloud explosions include gas explosions within buildings as well as outdoors (unconfined explosions). Fires constitute 35 % of the total damage. Apart from statistics also single accidents illustrate the damage potential of gas explosions such as the explosions which happened in Flixborough (2) in 1974, in Beek (3) in 1975 and on the offshore *rig Piper Alpha (4) in 1988. The major damage due to the latter accident was caused by a fire.* The escalation was, however, possible due to the consequences of an initial gas explosion.

All these accidents show that there it is very important to prevent gas explosions by reducing the risk of accidental releases, formation of explosive clouds and ignition. The loss experience, however, also shows that preventive measures are not sufficient. The frequency of gas explosions is still not sufficiently low to rely on preventive measures only. Therefore one has to build in a last barrier against gas explosions into the facilities, i.e. structural measures. The three accidents mentioned above all show that adequate prediction of the structural consequences of gas explosions would have prevented fatalities (many fatalities occurred in the control rooms which were not built strong enough to withstand explosions) and domino-effects (subsequent fires).

The damage potential of gas explosions is mainly due to a significant increase of the energy release rate resulting from an interaction of turbulence generated ahead of the flame and the flame: In an area with process equipment, piping, etc. the flame may accelerate to several hundred meters per second during a gas explosion. Upon ignition the flame will consume unburnt gas. The resulting combustion products will expand. This expansion can be up to 8-9 times the initial volume. Unburnt gas is therefore pushed ahead of the flame and a flow field will be generated. At equipment turbulence will be generated. When the flame propagates into this turbulent flow field, the burning rate will increase dramatically. This increased burning rate, i.e. an increase of the production rate of combustion products, will further increase the flow velocity and turbulence ahead of the flame, etc..

This mechanism of flame acceleration due to repeated obstacles constitutes a strong positive feedback loop. This loop is shown in Figure 1.

Predictive tools for explosion effects should reckon with this positive feedback mechanism and should also realize the sensitivity of this process to several scenario parameters such as ignition point location, gas cloud size, gas type, gas concentration, equipment location, equipment density and shape and partial confinement due to walls. As stated in (5) only numerical models provide the framework necessary to describe closely all of the fluid dynamic and combustion processes relevant to explosion prediction. During the past 13 years research has been carried out at Christian Michelsen Research to develop such a simulator. Lately the most recent version of this simulator FLACS-93 was issued (6).

The development of these tools necessitates experimental data regarding the influence of several of the scenario parameters mentioned above. One of these scenario parameters is the gas type. In the past CMR performed a comprehensive experimental programme to investigate the turbulent explosion properties of single component gases for both stoichiometric mixtures (7-9) and non-stoichiometric mixtures (10). Both offshore and onshore process streams, however, involve in many occasions mixtures of gases. An important example is natural gas which is a mixture of methane, ethane and some higher hydrocarbon gases. It was therefore decided to start investigations into the turbulent explosion properties of mixtures of gases. These investigations were performed on small- and large-scale using a wedge-shaped vessel.

The present paper reviews the main findings of these experiments.

EXPERIMENTAL ARRANGEMENT

Small-scale set-up

The small-scale version of the wedge-shaped explosion vessel is shown in Figure 2. The vessel is 1 m long and 0.125 m high giving a volume of approximately 18.5 l. The top angle of the vessel is 17°. The top plate of the vessel can either be solid or perforated. For the present experiments it was chosen to be a solid transparent plexiglass plate to allow for filming. Ignition was effected in the sharp end of the vessel using an electric spark. The other far end of the vessel was open to allow for venting. To cause flame accelerations during the explosions 5 baffle-like obstructions were included, blocking 16 %, 33 % or 50 % of the cross-section of the vessel.

The combination of sharp-edged obstacles, a solid top plate and ignition far away from the vent opening will cause the onset of a very positive feedback mechanism causing very intense turbulence conditions as desired (compare Figure 1).

Gas mixtures were prepared using a recirculation system . Gases were sucked from the vessel near the open end of the vessel and introduced in the vessel again close to the ignition point. To prepare gas mixtures and to mix them with air, the various gas components were added to the recirculating flow.

The gas mixtures were analysed using an infrared gas analyser (Binos, Leybold-Heraeus) and a gas chromatograph. The monitoring occurred continuously up to approximately 2 minutes before the moment of ignition at three positions in the recirculation system.

Figure 2 shows the experimental set-up including the location of instrumentation.

The diagnostics during the small-scale investigations consisted of pressure measurements at three locations and the measurement of moment of flame arrival at five locations in the vessel. The positions of these diagnostics have been indicated in Figure 2.

To register the measuring data a data-acquisition system was used consisting of a 1 MHz 12 channel datalogger.

Large-scale set-up

The dimensions of the large-scale set-up are 10 times the dimensions of the small-scale set-up. The volume of the vessel is 18.5 m^3. The length is 10 m and the height is 1.25 m. Inside the vessel different types (i.e. round cylinders, flat plates and boxes) and numbers of obstacles can be mounted. The top plate of the vessel can either be solid or perforated.

Figure 3 shows the set-up as used in the present set-up. The top plate was solid in all experiments. Ignition was always effected in the sharp-end of the vessel. The far end of the vessel was open. Five baffle-like obstacles were used blocking 33 % of the cross-section of the vessel.

Also in the large-scale experiments the gas mixtures were prepared using a recirculation system . To prepare gas mixtures and to mix them with air, the various gas components were added to the recirculating flow.

The gas mixtures were analysed using an infrared gas analyser (Binos, Leybold-Heraeus). The monitoring occurred continuously at three positions in the recirculation system.

The diagnostics during the large-scale investigations consisted of pressure measurements at five locations and the measurement of moment of flame arrival at 19 locations in the vessel. The pressure transducers are evenly distributed in the top plate of the vessel. The ionisation gauges were installed centrally between the top plate and the edge of the obstacles.

To register the measuring data a data-acquisition system was used consisting of a 1 MHz 12 channel datalogger.

Gas mixtures used in the experiments

The choice of the gas mixtures which were investigated was generally based on gas mixtures which can be encountered offshore. The following gas mixtures were used:

- dry natural gas (containing 85. 33 % methane, 11.76 % ethane, 0.94 % propane, 0.04 % i-butane, 0.06 % n-butane, 0.9 % nitrogen and 0.97 % CO_2)
- wet natural gas (containing 78. 95 % methane, 9.91 % ethane, 6.01 % propane, 0.75 % i-butane, 1.56 % n-butane, 1.01 % C_5^+, 0.85 % nitrogen and 0.96 % CO_2)
- methane-ethane mixtures
- methane-propane mixtures
- a methane-hydrogen mixture (containing 48 % methane and 50 % hydrogen)
- propane-carbon dioxide mixtures

As the overview of the tested gas mixtures show only two component gas mixtures were prepared. The two types of natural gas and the hydrogen-methane mixture were supplied by Statoil and Norsk Hydro. To prepare optimal mixtures of two gas components with air, with respect to explosion effects, the optimal concentrations of the single gas components (C_{opt}) were used as reference values. The optimal concentrations of the single components are near-stoichiometric concentrations ($C_{opt} \approx C_{st}$). The concentration of the optimal mixture of the two-component gas mixture is then given by:

$$C_{opt}\,(\text{mixture}) = \chi . C_{opt}\,(\text{component } 1) + (1 - \chi)\, C_{opt}(\text{component } 2)$$

These optimal concentrations were checked during the experiments by varying the concentration slightly around these concentration. χ is a number between 0 and 1.

RESULTS AND DISCUSSION

Small-scale experiments

The results of methane-ethane mixtures are presented in Figures 4 and 5. Figure 4 shows the terminal flame speed as a function of the gas mixture composition for the three different blockage ratios that were studied. The terminal flame speeds are measured being an average value of the flame speed over a distance of 10 cm close to the open end of the small-scale set-up. The results show first of all that the terminal flame speeds increase with increase of the blockage ratio as known from several earlier investigations (8). The most important observation though is that the terminal flame speed decreases almost linearly with mixture composition (with increase of the amount of methane in the mixture). This observation accounts for all blockage ratios that were applied in these experiments.

In the Figure the two types of natural gas that were tested were included as well using the methane concentration as a reference parameter. For the sake of the Figure the remainder was assumed to be ethane. The results for these two gases appear to be consistent with the results of the methane-ethane mixtures for all three obstacle configurations. Figure 5 shows that also the pressure decreases almost linearly. According to calculations by Kuhl et al. (11) using a similarity solution the relationship between flame speed and pressure is exponential for cylindrical geometries assuming a plane flame. For a cylinder-symmetrical geometry as the wedge-shaped vessel one would therefore expect a more exponential decrease of pressure as a function of the mixture composition regarding the linear relationship between flame speed and mixture composition. This observation implies that the flame is not a plane flame but that the flame is highly deformated and that a part of the burning occurs in unburned gas pockets in between the obstacles. This has been illustrated in Figure 6.

For propane-methane-air mixtures similar trends were found. The maximum flame speed of pure propane-air mixtures considering a 33 % blockage ratio was 80 m/s (slightly lower than found for pure ethane) and the maximum overpressure that was generated for this situation amounts to 0.14 bar.

The results of the hydrogen-methane mixture are given in Table 1.

Table 1. Results for hydrogen-methane mixtures

	Pure methane (present work)		Methane/hydrogen (present work)		Pure hydrogen Bjørkhaug (12)	
Blockage ratio (%)	Overpressure (bar)	Flame speed (m/s)	Overpressure (bar)	Flame speed (m/s)	Overpressure (bar)	Flame speed (m/s
16	0.059	37.0	0.130	66.5	-	-
33	0.080	69.9	0.200	127.0	-	-
50	0.196	88.8	0.675	148.9	3.14	422.5

The table presents the results for pure methane and the hydrogen/methane mixture found in the present experiments. No experiments were performed using pure hydrogen but to allow for investigating the effect of mixture composition over the full range from pure methane to pure hydrogen mixtures results of experiments reported by Bjørkhaug (12) and performed in the same experimental set-up using pure hydrogen were adopted. The optimal mixture of 50 % v/v methane (assumed instead of the 48 % v/v in reality) and 50 % v/v hydrogen is a mixture of 0.25 C_{opt} (hydrogen) and $0.75C_{opt}$ (methane). Comparison of the results for the 50 % blockage ratio situation shows that also for these mixtures a linear relationship appears to exist considering flame speeds. The pressure-composition dependency though is non-linear reflecting the higher flame speeds that were obtained. According to the calculations by Kuhl et al. (11) the non-linearity should increase with an increase of flame speeds.

As the composition of the two types of natural gas indicates (section 2.3), there are several mixtures which contain small amounts of inert gases such as carbon dioxide and nitrogen. To investigate the influence of inert gases on the flame propagation in congested environments some tests were performed with mixtures of propane and carbon dioxide. In these tests the concentration of carbon dioxide was gradually increased in proportion to the propane concentration (25 %, 50 %, 100 %). Results are shown in Figure 7. The experiments show that only very large amounts of carbon dioxide (comparable to the concentration of the flammable gas) cause an effect on the maximum overpressures generated in the wedge-shaped vessel. This effect is caused due to reduction of the oxygen concentration. This will result in lower burning velocities. This has a direct effect on the flame speed development and therefore pressure development but in addition to that the lower reactivity of the mixture may more easily result in quenching in areas where very high intensity turbulence exists during the explosions. This will result in additional reduction of overpressures.

Large-scale experiments

Some of the experiments performed on small-scale were repeated on large scale. On large scale one would expect the same tendencies as found on small scale considering the effect of mixture composition on flame speed and pressure development. Flame speeds and maximum overpressures will be higher however, due to the effects of hydrodynamical instabilities which will affect the initial flame speed development on large scale (13) and due to the effect of turbulent length scale on the burning rate (14).

In general the large-scale results confirm the small-scale findings. Figure 8 shows flame speed-distance relationships measured for 4 tests using pure 9.8 % v/v methane-air mixtures. The results show that the flame trajectories initially are very much alike . The position of the obstacles at 2 m, 4 m etc. from the ignition source location can clearly be recognized due to the local flame accelerations at these positions. During the later stages of flame propagation the repeatibility appears to decrease and a relatively large scatter of terminal flame speeds is found. This scatter is found again in the maximum overpressures that are measured. Figure 9 presents the results of maximum overpressures measured for several methane-propane mixtures. The non-linear curve shows the best fit of the pressure results reflecting similar small-scale results. If dry natural gas is represented by a methane-propane mixture representing methane as methane and all other gases as propane one ends up at a reasonable agreement between the pressures generated by methane-propane mixtures and the single result for natural gas.

A comparison with small-scale experiments shows that the pressures generated in the large-scale experiments are an order of magnitude higher. This reflects flame speeds which were approximately 5 times as high on large scale than they were on small scale. As mentioned before the higher flame speeds are caused by the effects of hydrodynamical instabilities and the effect of turbulent length scale on the burning rate. The higher flame speeds will cause a stronger non-linearity in the pressure-mixture composition relationship than found on small scale. This is in agreement with the results of the calculations published by Kuhl et al. (11).

FURTHER DISCUSSION

The results of the present experiments seem to suggest that there is a linear relationship between mixture composition of two component gas mixtures and turbulent flame speeds generated in a wedge-shaped vessel. This seems to suggest that the only parameters determining the increase of the burning rate when changing the mixture composition are the laminar burning velocity S_L and the expansion ratio ρ_u/ρ_b (ratio of densities of unburnt gas and burnt gas respectively) being in agreement with earlier observations on the effect of reactivity on flame propagation in congested environments (15)

Based on experimental data gathered by Abdel-Gayed et al. (14), Bray (16) suggested the following relationship for turbulent burning velocities S_T:

$S_T = 0.875\ K^{-0.392}\ u'$ where $K = 0.157\ (u'/S_L)^2 (\nu/u' l_t)^{0.5}$
u' = turbulence intensity
S_L = laminar burning velocity
ν = kinematic viscosity
l_t = turbulence length scale

If we assume the kinematic viscosity ν to be constant : $\nu = 2.10^{-5}\ m^2/s$ this relationship becomes:

$$S_T = 15.0\ S_L^{0.784} u'^{0.412} l_t^{0.196}$$

Using this relationship it is investigated whether a linear relationship between flame speed and mixture composition could be expected. It is assumed that the turbulence intensity in the wake of an obstacle is directly proportional to the flow speed ahead of the flame in front of that obstacle. The turbulence length scale is assumed to be constant throughout the vessel. The effect of flame folding and the effect of hydrodynamical instabilities have been neglected.

Thus it is found:

$$V_f = 88\ \rho_u/\rho_b^{1.68}\ p^{1.68} S_L^{1.32} l_t^{0.32}$$

where p = constant describing relationship
between turbulence intensity in the
wake of an obstacle and the main
flow speed

The absolute flame speeds which are found in this way are a factor of approximately 1.5-4.5 (regarding several blockage ratios) lower than one would expect for the small scale experiments and a factor of approx. 7 (BR= 0.33) lower than found for the large scale situation. This illustrates the importance of the effect of flame folding and the effect of hydrodynamical flame instabilities on large scale. Nevertheless as the flame folding will be directly proportional to the turbulence intensity and as the effect of hydrodynamical instabilities on flame speeds are known the relationship allows for investigating the relative effects of scale and reactivity on flame speeds. If we assume that the laminar burning velocity and the expansion ratio of 2 component mixtures increase linear with the mixture composition the relationship shows that one would expect a close to linear relationship of flame speeds thus confirming the present results. With respect to scaling the relationship shows one would expect an increase of a factor of 2 for the flame speeds on large scale in comparison to the small-scale results. A factor of 5 was measured implying that hydrodynamical instabilities would cause an additional flame speed increase on large scale by a factor of 2.5. Hydrodynamical instabilities are important for flame propagation up to the first obstruction. On large scale this distance is approximately 2 m and one should at such a distance indeed expect an increase of flame speeds due to such instabilities by a factor of 2-3 and therefore the relationship appears to describe the effects of scaling correctly also.

The discussions mentioned above justifies the inclusion of the Bray´s combustion model in the new FLACS-simulator (6).

CONCLUSIONS

Explosion experiments carried out in a wedge-shaped vessel with obstacles to investigate the turbulent explosion properties of mixtures of gases shows that:

- the turbulent burning rate of mixtures of two gases lies in between the turbulent burning rate of the pure gases
- the total turbulent combustion rate of a mixture of two gases increases non-linearly with the laminar burning rate of this mixture
- the adding of small amounts of carbon dioxide to a flammable gas has hardly any effect on its turbulent explosion properties
- he turbulent burning rate of natural gas can be represented satisfactorily by the turbulent burning rate of mixtures of methane and ethane or methane and propane
- flame speeds derived from measurements of the moment of arrival of the flame tip is not necessarily representative for the total burning rate of gas mixtures in a congested environment

ACKNOWLEDGEMENT

This work has financially been supported by BMFT, BP, Conoco, Elf, Esso, Gasunie, Gaz de France, HSE, Mobil, Norsk Hydro, NPD, Phillips and Statoil. The authors acknowledge the technical support of Mr. A. Lauritzen, Mr. J.F. Beuvin and Ms. E.K. Sæthre.

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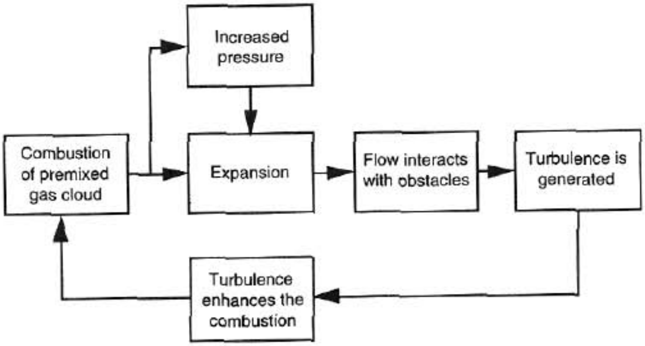

Figure 1. Positive feedback loop causing flame acceleration due to turbulence.

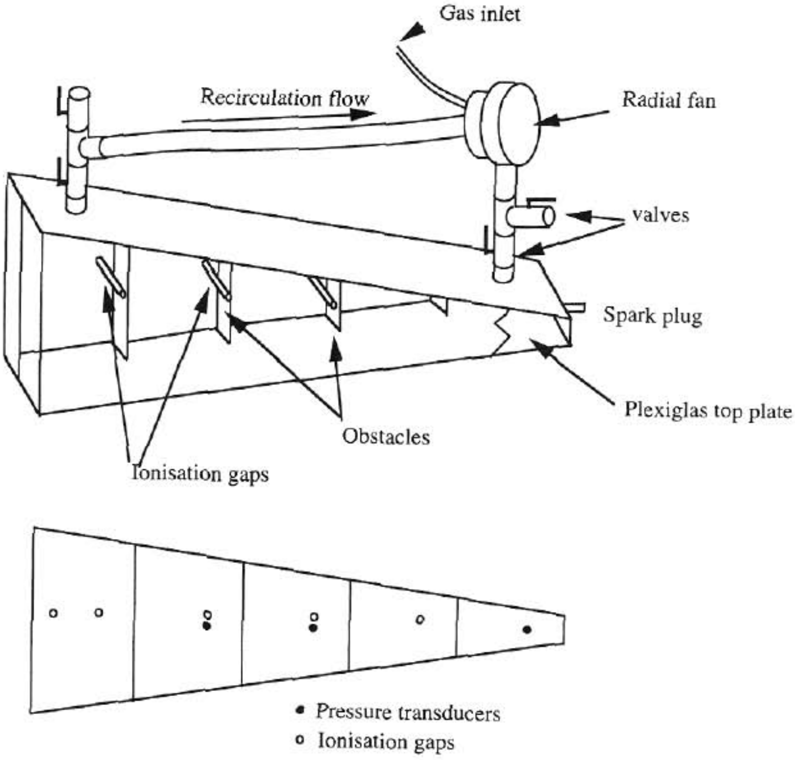

Figure 2. Small-scale experimental set-up showing the recirculation system and instrumentation.

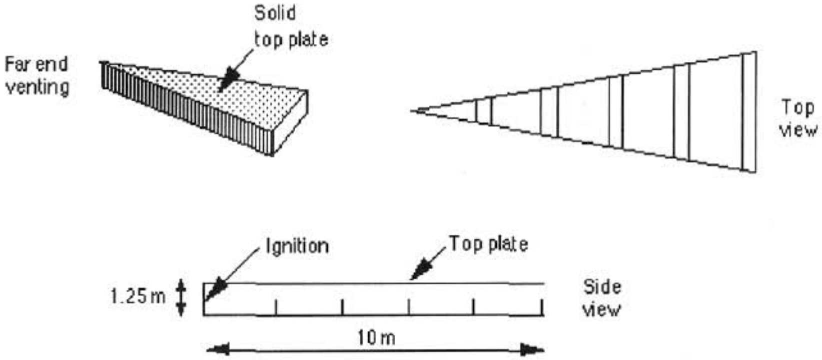

Figure 3. *Large-scale wedge-shaped explosion vessel.*

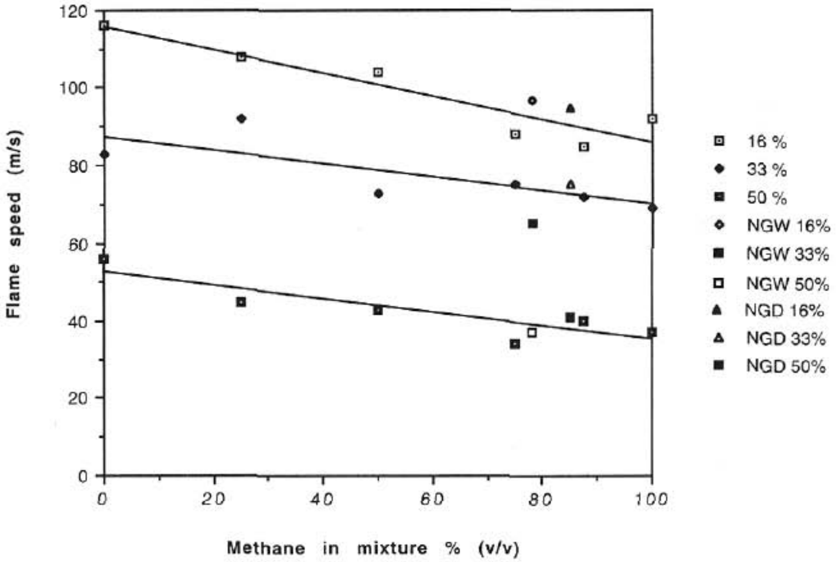

Figure 4. *Flame speed as a function of mixture composition for methane-ethane mixtures using obstacles blocking 16 %, 33 % and 50 % of the cross section (NGW = natural gas wet; NGD = natural gas dry)*

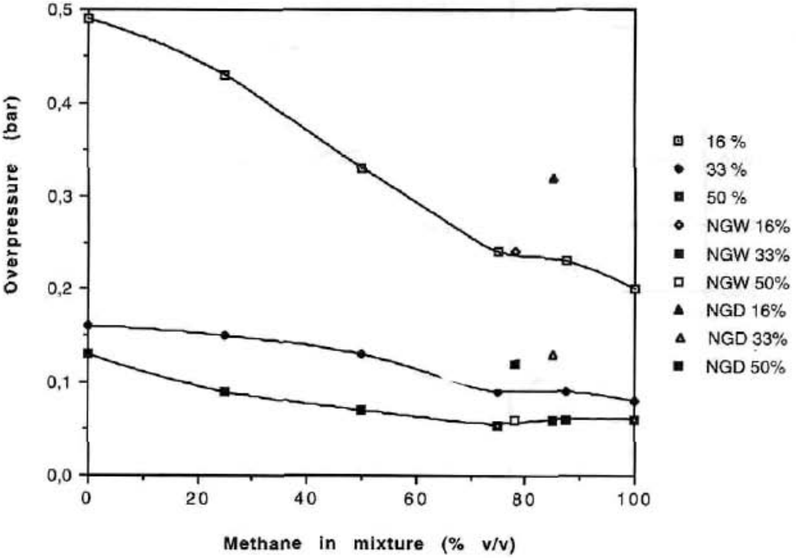

Figure 5. Maximum overpressure as a function of mixture composition for methane-ethane mixtures using obstacles blocking 16 %, 33 % and 50 % of the cross section (NGW = natural gas wet; NGD = natural gas dry)

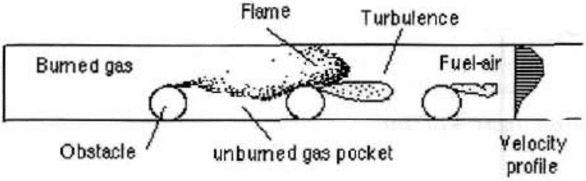

Figure 6. Moment of flame propagation in wedge-shaped explosion vessel.

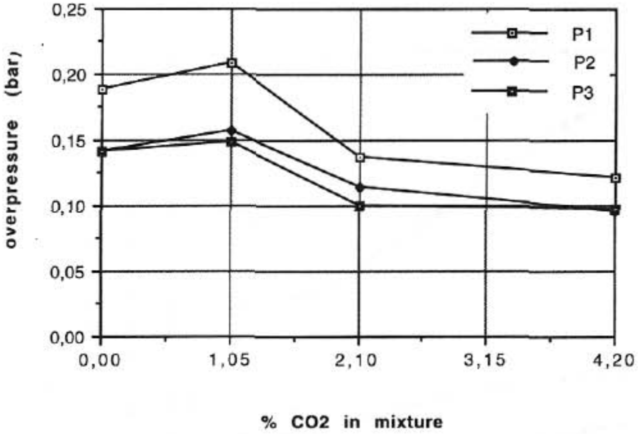

Figure 7. Influence of carbon dioxide on explosion propagation in a small-scale wedge-shaped vessel.

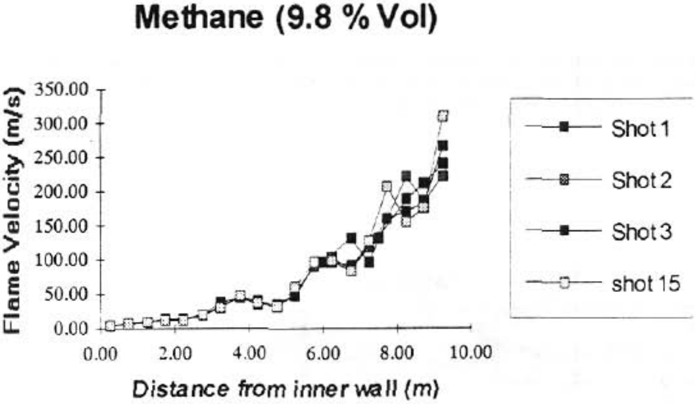

Figure 8. Flame speed-distance relationship measured for pure methane-air mixtures.

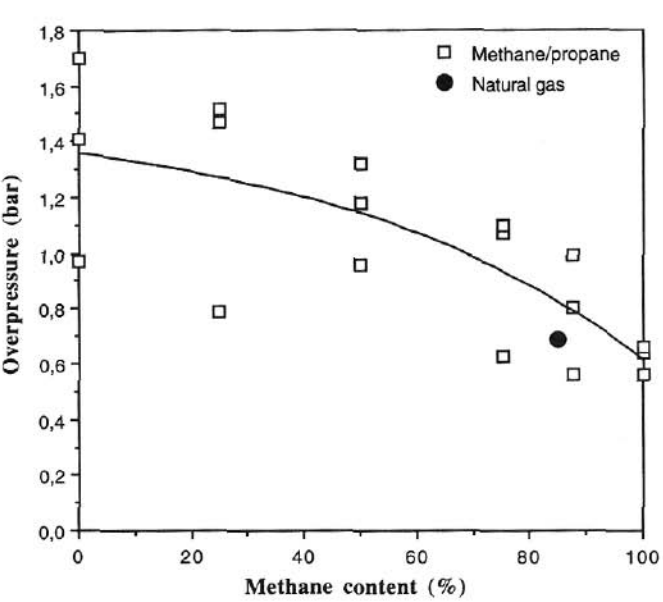

Figure 9. Maximum overpressure as a function of mixture composition for methane-propane mixtures using obstacles blocking 33 % of the cross section (of the large-scale wedge shaped vessel.