EFFECT OF CARBON DIOXIDE, ARGON AND HYDROCARBON FUELS ON THE STABILITY OF HYDROGEN JET FLAMES

Wu, Y[1], Al-Rahbi, I. S.[1] , Lu, Y[1]. and Kalghatgi, G. T.[2]
[1]Department of Chemical and Process Engineering, University of Sheffield, Sheffield S1 3JD, UK
[2]Shell Global Solutions (UK), Cheshire Innovation Park, P.O. Box 1, Chester CH1 3SH, UK

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ABSTRACT

Experimental studies were carried out to examine the lift-off and blow-out stability of H_2/CO_2, H_2/Ar, H_2/C_3H_8 and H_2/CH_4 jet flames. The experiments were carried out using a burner with a 2mm inner diameter. The flame structures were recorded by direct filming and also by a schlieren apparatus. The experiments showed that the four gases affected the lift-off and blow-out stability of the hydrogen differently. The experiments showed that propane addition to an initially attached flame always produced lifted flame and the flame was blown out at higher jet velocity. The blow-out velocity decreased as the increasing in propane concentration. Direct blow-off of hydrogen/propane was never observed. Methane addition resulted in a relatively stable flame comparing with the carbon dioxide and propane addition. Comparisons of the stability of H_2/C_3H_8, H_2/CH_4 and H_2/CO_2 flames showed that H_2/C_3H_8 produced the highest lift-off height. Propane is much more effective in lift-off and blow out hydrogen flames. The study carried out a chemical kinetic analysis of H_2/CO_2, H_2/Ar, H_2/C_3H_8 and H_2/CH_4 flames for a comparison of effect of chemical kinetics on flame stability.

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1 INTRODUCTION

The lift-off and blow-out stability parameters have significant meaning in the diffusion flame modelling and also have strong implication in safety consideration and hazard assessment. Hydrogen fuel has great advantages in environment implication and it is perceived as an ideal energy carrier for a clean and sustainable energy future. For the foreseeable future, hydrogen will co-exist with types of hydrocarbon fuels in both the public and domestic environments. Therefore study of effects of hydrocarbon fuels on hydrogen has significant meanings in usage and transport hydrogen. This study is aimed at investigation of the factors affecting the stability of hydrogen jet flame and the effect of hydrocarbon fuels on the stability of hydrogen jet flames.

2 THE EXPERIMENTAL STUDY

The experiments were carried out using a burner with a 2 mm inner diameter. The burner was fitted with flow settling chamber and flow straightening device. The gasses were introduced from compressed gas bottles through flow meters and were mixed before channeling into the settling chambers. The visual characteristics of the flames of pure hydrogen were very different from H_2/C_3H_8, H_2/CH_4 and H_2/CO_2 flames. The pure hydrogen jet flames were almost invisible. H_2/CO_2 flames and H_2/CH_4 were blue. The H_2/C_3H_8 flames resembled characteristics of propane ones and appeared in blue in the base of the flame, but bright yellow in the main combustion zone. To visualise the flames and establish the lift off height of the flames, both schlieren technology and direct digital photography technology were used to capture the flame images.

A positive-negative-grid schlieren system was constructed for the rig and this was mainly used to visualise hydrogen, hydrogen/carbon dioxide flames. All flame images were captured using digital camera and processed using computer graphic packages.

3 EXPERIMENTAL RESULTS

3.1 Stability of Pure Hydrogen Jet Flames

The experimental results showed that pure hydrogen jet flame was very stable. The lift-off velocity measured for pure hydrogen flame was as high as 738 m/s. The hydrogen flame lift-off height was measured by initially increasing the flowrate at regular intervals until the exit velocity reached 1294 m/s where Ma=1, then reducing the flowrate until the flame re-attached the burner. Figure 1 plotted hydrogen flame lift-off height data obtained from the present study and some experimental data obtained by Kalghatgi [1], Cheng and Chiou [2] and Al-Rahbi [3]. The results showed that the lift-off height increased linearly with the jet velocity. The measured lift-off heights from different studies using different burner diameter are in reasonable agreement between the jet velocity 750 m/s to 1200 m/s. The lift-off velocity for hydrogen jet flame was between 500 m/s to 700 m/s depending on the burner and jet diameter. There was some diversity in the lift-off height in the transition from attached to completely lifted flame. Above 1200m/s, hydrogen jet reached supersonic flow, the errors in jet velocity value would increase causing diversity in the flame lift-off height. Blow out of pure hydrogen jet flame has never been achieved in the present study.

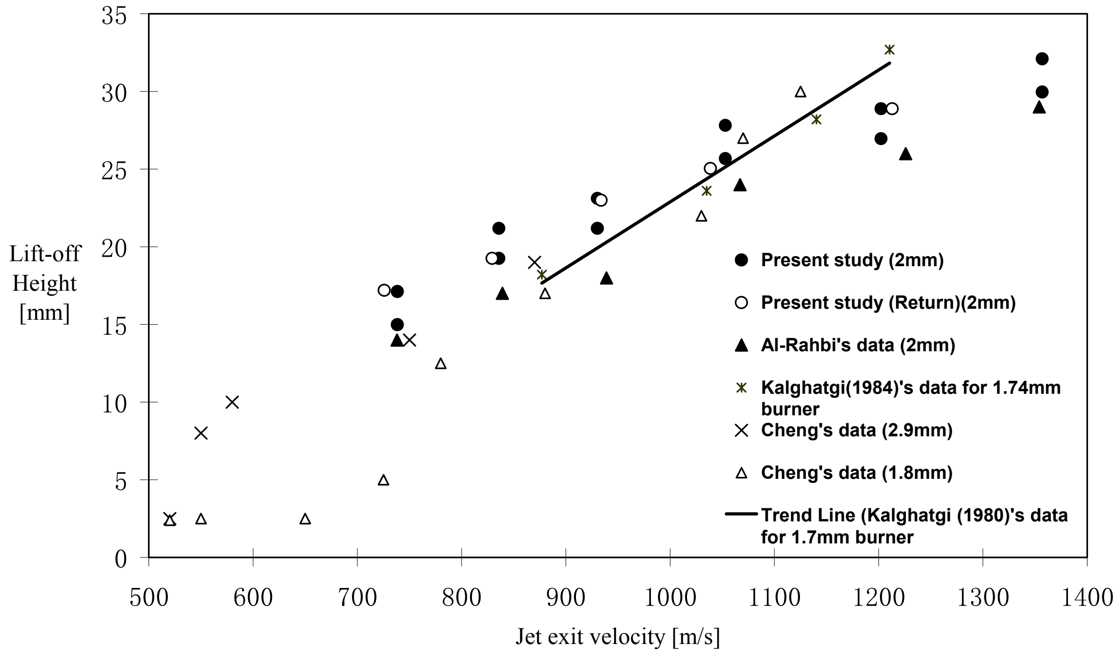

Figure 1. Comparison of experimental measured lift-off height of pure hydrogen jet flames against the jet velocity.

Figure 2 presents a comparison of the measured lift-off heights and that predicted using the correlation given by Kalghatgi [1]. The predictions were made using the maximum laminar burning velocity value, S_L=3.06 m/s, as given in Kalghatgi [1] and also using the Stoichiometric burning velocity S_L=2.25 m/s.

It was shown that the measured lift-off heights are approximately a factor of 2 higher than those predicted by Kalghatgi's correlation when using the maximum laminar burning velocity value. If the burning velocity of stoichiometric H_2-Air is used in Kalghatgi's equation, then the correlation by Kalghatgi fits much better with the experimental data as illustrated in Figure 2. Pitts [4] also observed this deviation when using Kalghatgi's formula.

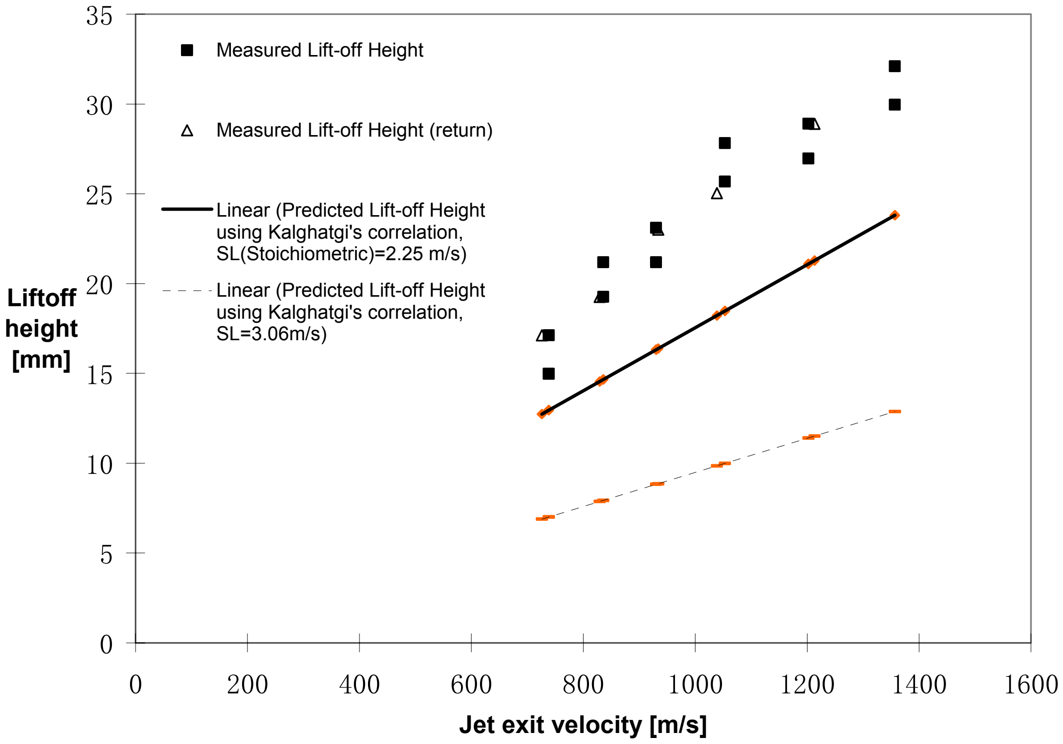

Figure 2. Comparison of experimental measured lift-off height with the prediction using correlation from Kalghatgi (1984).

In Kalghatgi's analysis it was assumed that the flame was stabilized when the maximum laminar burning velocity was obtained. For most hydrocarbon fuels, the maximum S_L occurs near to stoichiometric equivalence ratio. Therefor, the stoichiometric burning velocity was usually used for calculation of the lift-off height for hydrocarbon fuels when using Kalghatgi's correlation. However, for H_2 the maximum burning velocity occurs on the rich side at an equivalence ratio of around 1.83. At the stoichiometric concentration, the laminar burning velocity for H_2 is about 2.25 m/s as was shown by Law [5]. The difference between H_2 and hydrocarbon fuels in terms of S_L revealed a grey area in the premixed flame propagation theory for stabilization mechanism of lifted jet flames. That is whether a lifted flame base exists at the stoichiometric contour or at the location where the maximum burning velocity is attained.

Studies aiming to clarifying this difference are quite rare in the literature. The experimental work of Tacke *et al.* [6] on undiluted H_2 and H_2-N_2 jet flames issuing into co-axial laminar flow of air showed that the lifted H_2/Air flame was stabilized where the fuel mixture fraction was around 0.0215. The DNS prediction of lifted H_2 jet diffusion flames, that were recently carried out by Mizobuchi *et al.* [7], have also shown that the flame was stabilized by the leading edge flame and that the S_L corresponding to the local mixture was about 2 m/s.

The two studies therefore suggest that the H_2 flame base is stabilized close to the stoichiometric contour, where S_L is about 2 m/s. The stoichiometric mass fraction that corresponds to this burning velocity is 0.028.

3.2 Effect of CO_2 Addition on the Stability of H_2 Jet Flames

Experimental tests were conducted to examine the effect of CO_2 addition on the stability of an initially attached and also an initially lifted H_2 flame. The experimental results showed that when CO_2 was added to an attached H_2 flame, two flame stability regimes were identified. There was a critical CO_2 concentration. If the CO_2 concentration was greater than 6.4 %, addition of CO_2 produced a stable attached flame leading to direct blow off at high velocities. If CO_2 concentration was less than 6.4 %, CO_2 addition produced a lifted flame and then the flame was blown out at higher velocities.

Experiments were also were carried out to examine the effect of CO_2 addition on lifted H_2 flames. It was shown that the CO_2 addition increased the lift-off height by nearly two times the initial lift-off height of pure H_2 flames.

3.3 Effect of C_3H_8 Addition on the Stability of H_2 Jet Flames

Experimental tests were carried out to study the effect of C_3H_8 addition on an initially attached and an initially lifted H_2 jet flame. C_3H_8 addition to an attached H_2 flames always produced lifted flames, which were blown out at high jet velocities. Direct flame blow off was not observed. C_3H_8 addition to lifted H_2 flames increased the lift-off height by nearly 2.6 times before blow out was observed. The blow out occurred at a C_3H_8 concentration of around 4 to 5 %.

3.4 Effect of CH_4 Addition on the Stability of H_2 Jet Flames

Test results demonstrated that effects of CH_4 addition were different from C_3H_8 addition. Similar to the effect of CO_2 addition, there were two flame stability regimes when CH_4 was added into initial attached hydrogen jet flames. If the CH_4 concentration was greater than 20%, flame would remain attached until blow-off at high velocity. If the CH_4 concentration is less than 20%, CH_4 addition to an initial attached hydrogen jet flame can produce lifted flame.

4. DISCUSSIONS

4.1 Lift-off Height

Figure 3 shows a comparison of the lift-off height of H_2/C_3H_8, H_2/CH_4 and H_2/CO_2 flames. The addition of the CH_4, CO_2 and C_3H_8 always increased the liftoff height of the initially lifted hydrogen diffusion flames. Addition of C_3H_8 produced the highest lift-off height. For the same jet exit velocity and same concentration of the diluents, the addition of C_3H_8 produced liftoff height around 15 to 30% higher than that of the addition of CO_2. For the similar amount of diluent addition, at the same jet exit velocity, the addition of CH_4 produced liftoff height lower than that of the addition of C_3H_8. At jet exit velocity higher than around 1000 m/s, the liftoff height of flames with CH_4 addition are shown to be higher than that with CO_2 addition. It might be predicted that at the high concentration of diluent (potentially be higher than 10%), the addition of CH_4 would produce higher lifted flames than that of CO_2.

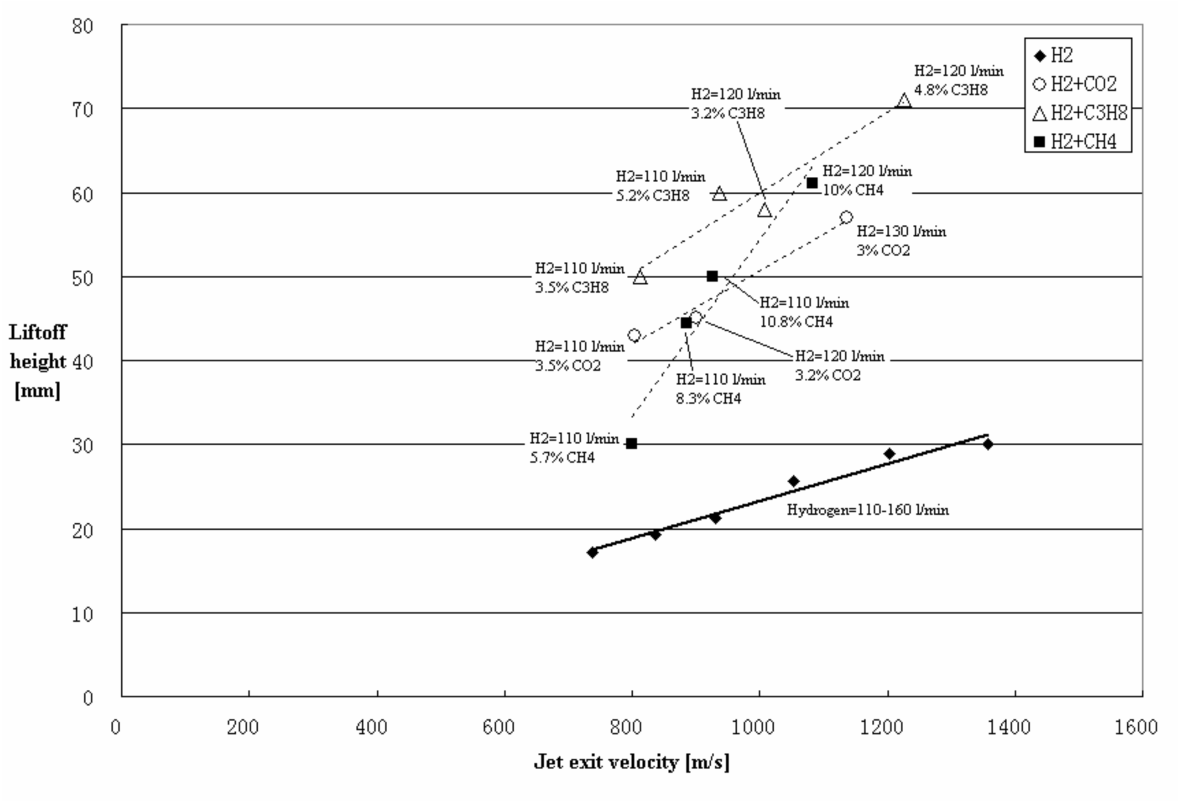

Figure 3. Comparison of the lift-off height of H_2/C_3H_8, H_2/CH_4 and H_2/CO_2 flames.

4.2 Lift-off Velocity

A comparison of effect of propane addition and methane additions on lift-off velocity is shown in Figure 4. The results showed that for the same concentration, methane addition required a much higher velocity for lift-off to occur than for propane addition.

4.3 Blow-out and Blow-off Velocity

Effects of C_3H_8, CH_4 and CO_2 additions on blow-out or blow-off velocity were very different. Addition of C_3H_8 produced lifted flame and then flame was blow-out at higher velocity. Addition of C_3H_8 didn't produce direct blow-off . CO_2 additions could produce lifted flame if the CO_2 concentration is low. At high CO_2 concentration, flame would remain attached until blow-off. Therefore for H_2/CO_2 flames both blow-out and blow-off velocities were obtained. For H_2/CH_4 flames, only blow-off at high concentration was obtained. The blow-out flame conditions were not tested yet in present study. Figure 5 plotted the blow-out and blow-off velocities against the addition concentration. It was shown that for the same jet velocity, CH_4 required much higher concentration to produce blow-off than CO_2 and C_3H_8. It was also showed that at high concentration (about 16%), C_3H_8 is most efficient in blow-out the flame and CH_4 is the least efficient agent to produce blow-out of hydrogen flame.

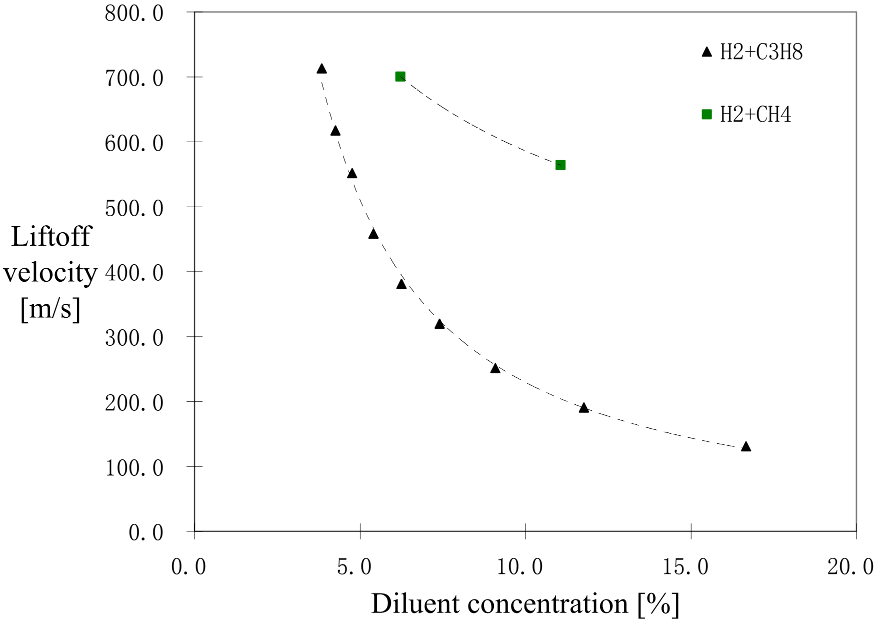

Figure 4. Comparison of lift-off velocity H_2/C_3H_8 and H_2/CH_4 flames

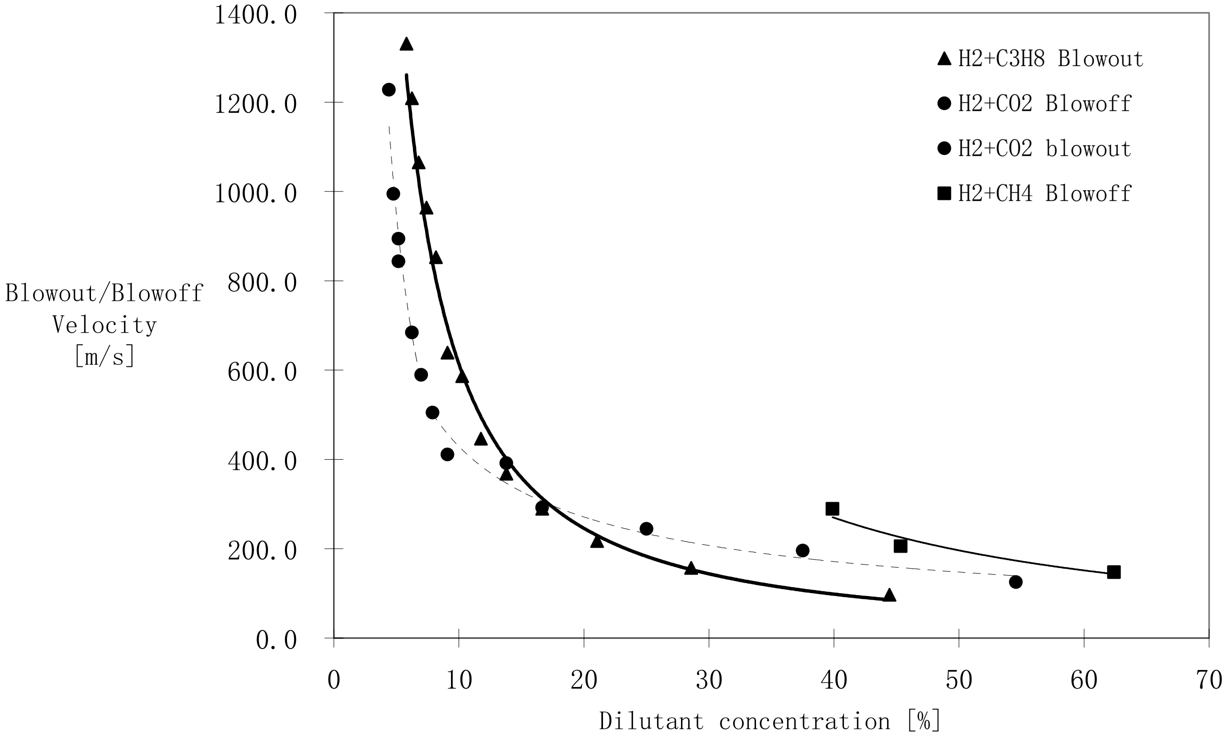

Figure 5. Comparison of blow-out and blow-off velocity of H_2/C_3H_8, H_2/CH_4 and H_2/CO_2 flames.

4.4 Laminar Burning Velocity

To analyse the experimental results, a detailed study and comprehensive literature review on the effect of propane and CO_2 addition on the burning velocity of hydrogen flames was carried out. Some limited research has been carried out on the effect of additive on the burning velocity of hydrocarbon fuel. The effect of CO_2 addition on the burning velocity of methane and propane has been well studied. Yumlu [8] experimentally studied the effect of additives on the burning speed of fuel-air mixtures using a circular burner. For adding inert gas, an equation for the burning velocity of the mixture, $S_{u,m}$, was derived as:

$$S_{u,m}^2 = (1-\alpha)^{1/2} S_u^0 \exp[-E/R(1/T_b - 1/T_b^0)] \tag{1}$$

Where α is the mass fraction of the additive, T_b^0 is the adiabatic flame temperature and E is the activation energy. However the effect of adding combustible gas to the fuel-air mixture was not well understood and there was very limited experimental data available on the laminar burning velocity of hydrogen/propane mixture. Leason [9] experimentally studied the effect of various additives including hydrogen on the burning velocity of propane-air mixture. Though this work covered an extended range of equivalence ratio, it was limited to hydrogen concentrations in the H_2-C_3H_8 mixtures between 5 and 30%. Hydrogen was the dominant fuel with small amount propane addition; therefore Leason's data were not very useful in our study. Milton and Keck [10] made a further study on the effect of hydrogen addition on the burning velocity of methane-air and propane-air mixtures at the stoichiometric ratio. Figure 6 shows a comparison of the effect of adding CO_2 on the burning velocity based on Yumlu's equation, the effect of adding propane on the burning velocity based on the experimental data from Milton and Keck and effect of adding methane based on data from Choudhuri, [11]; Law and Kwon [12].

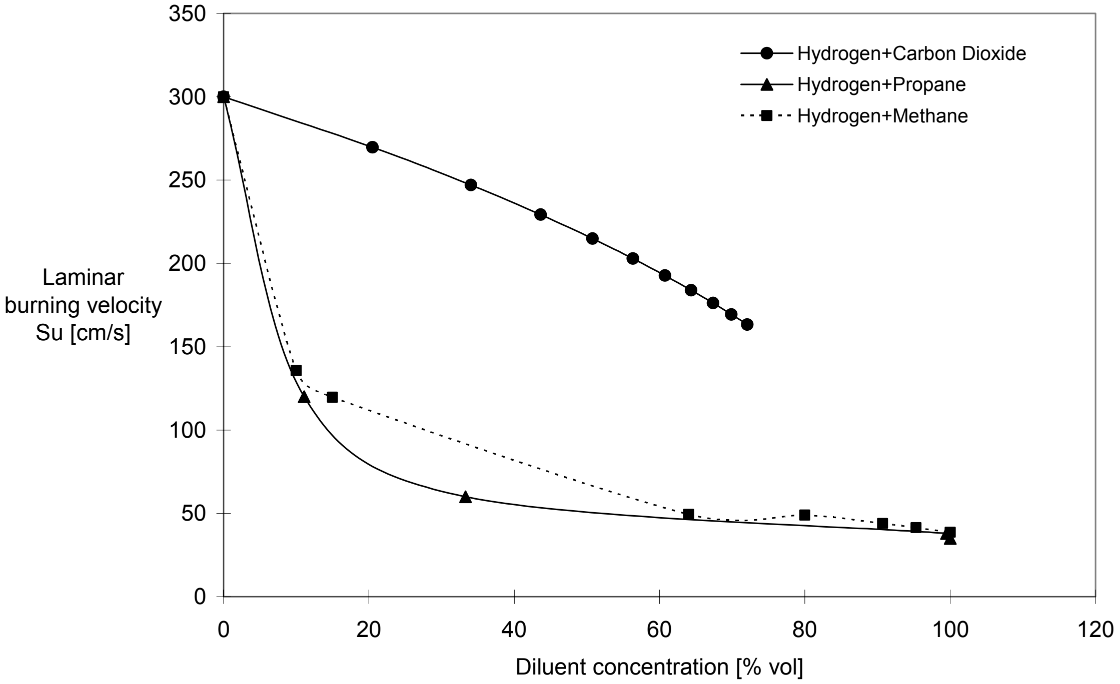

Figure 6. The laminar burning velocity of H_2/C_3H_8, H_2/CH_4 and H_2/CO_2 flames

It was shown that the reduction on the burning velocity by adding propane was much more significant and greater than by adding CO_2. Adding a small amount of propane reduced the burning velocity significantly. If the percentage of propane was greater than 30%, the burning velocity of the mixture was equal to that of pure propane fuel. The analysis revealed that it was propane, rather than hydrogen that seemed to be the dominant element on the burning velocity of the mixture.

An examination of the chemical reactions that are involved in the H_2-CO_2 and H_2-C_3H_8 mixtures was carried out and the kinetic mechanism for CO/CO_2, H_2/O_2 reactions and C_3H_8 destruction is provided in Table 1.

Table 1: Kinetic mechanism for CO/CO_2, H_2/O_2 reactions and C_3H_8 destruction from Peters and Rogg (1993).

No	**Reaction**	**A [mole, cm, s]**	**n**	**E [kJ/mole]**
	CO/CO_2 Mechanism			
1	$CO + OH \rightarrow CO_2 + H$	4.400E+06	1.5	-3.10
2	$CO_2 + H \rightarrow CO + OH$	4.956E+08	1.5	89.76
	H_2/O_2 Chain Reactions			
3	$O_2 + H \rightarrow OH + O$	2.000E+14	0.00	70.30
4	$OH + O \rightarrow O_2 + H$	1.568E+13	0.00	3.52
5	$H_2 + O \rightarrow OH + H$	5.060E+04	2.67	26.30
6	$OH + H \rightarrow H_2 + O$	2.222E+04	2.67	18.29
7	$H_2 + OH \rightarrow H_2O + H$	1.000E+08	1.60	13.80
8	$H_2O + H \rightarrow H_2 + OH$	4.312E+08	1.60	76.46
9	$OH + OH \rightarrow H_2O + O$	1.500E+09	1.14	0.42
10	$H_2O + O \rightarrow OH + OH$	1.473E+10	1.14	71.09
	C_3H_8 Consumption			
11	$C_3H_8 + H \rightarrow n\text{-}C_3H_7 + H_2$	1.300E+14	0.00	40.60
12	$C_3H_8 + H \rightarrow i\text{-}C_3H_7 + H_2$	1.000E+14	0.00	34.9
13	$C_3H_8 + O \rightarrow n\text{-}C_3H_7 + OH$	3.000E+13	0.00	24.1
14	$C_3H_8 + O \rightarrow i\text{-}C_3H_7 + OH$	2.600E+13	0.00	18.7
15	$C_3H_8 + OH \rightarrow n\text{-}C_3H_7 + H_2O$	3.700E+12	0.00	6.9
16	$C_3H_8 + OH \rightarrow i\text{-}C_3H_7 + H_2O$	2.800E+12	0.00	3.6

A = Pre-exponential factor; n = Temperature exponent; E = Activation energy

The CO/CO_2 reaction with O and OH is given by reactions 1 and 2. Reactions 3 to 10 provide the H_2/O_2 mechanism. The destruction of C_3H_8 by H, O and OH radicals is given by reaction 11 to 16.

The kinetic mechanism showed that CO_2 is not completely inert when mixed with H_2. CO_2 actually participates in the so-called gas shift reaction in which it combines with H radicals to form CO and OH, as given in the reactions 1 and 2 (Turns [13]).

From the reactions 11 to 16 it is shown that C_3H_8 breaks down to form lighter hydrocarbon radical (C_3H_7) and water. The lighter C_3H_7 molecule disintegrates to smaller hydrocarbon molecules via other reactions, which for the purpose of simplicity are not shown here. Since the three radicals (i.e. H^+, O^{-2}, OH^-) are of vital importance for the chain propagation reactions of H_2 represented here by reactions 3 to 10, it is considered that C_3H_8 addition to H_2-Air mixture would therefore hinder the reaction of H_2 with air as C_3H_8 would potentially act as a sink for the H, O and OH free radicals.

This analysis of the chemical kinetics does in principle explain the dominance of C_3H_8 combustion in H_2-C_3H_8 mixtures and the rapid reduction of the laminar burning velocity of H_2 when mixed with C_3H_8. It is also noted that both CO_2 and C_3H_8 can react with H radical; however the rate of C_3H_8 reaction with H is 10^6 faster than the case with CO_2 as illustrated by reaction 11 and 2, respectively.

The effect of CO_2 was compared with that of Ar, which is an inert that does not participate in the combustion reaction. For this reason, Ar was selected as the bench- mark in this study. Experimental results showed that CO_2 affected the H_2 flame stability in the same way as Ar. This further suggests that CO_2 acted more like an inert when added to H_2 jet flames.

4.5 Stoichiometric Mass Fraction

Unlike CO_2, the addition of C_3H_8 and CH_4 also changes the mixture's stoichiometric fuel/ air ratio. To illustrate the effect of C_3H_8 addition on the stoichiometric mass fraction, Figure 7 presents the variation of stoichiometric fuel mass fraction of the H_2-C_3H_8 blend as a function of C_3H_8 addition. It can be seen that at concentrations greater than 50 %, the fuel mass fraction approaches that of pure C_3H_8 of 0.06. The jet flame therefore becomes richer as propane is added. The effect of adding CH_4 was not less than the one of adding C_3H_8 .

Previous studies by Kalghatgi [14] and Broadwell [15] showed that for a given fuel the blow out velocity is proportional to S_L^2. In this study, comparing two different fuels, the experimental results showed that the blow out velocity for H_2-C_3H_8 is higher than that of H_2-CO_2 jet flames, although the H_2-C_3H_8 blends have a lower burning velocity than H_2-CO_2.

It was indicated in the study of Kalghatgi [1] that flame blow out occurs when the lift-off height is about 65 to 75 % of H (along the jet axis) where the stoichiometric fuel mass fraction is expected to occur. The flame base was not expected to exist beyond this distance. The stoichiometric fuel mass fraction in air (Y_{ST}) of H_2-C_3H_8 increases with C_3H_8 concentration. This implies that the jet fluid becomes richer with C_3H_8 addition and therefore requires a longer distance to attain stoichiometric concentration. Thus H_2-C_3H_8 mixtures require higher exit velocity to cause flame blow out than H_2-CO_2 mixtures.

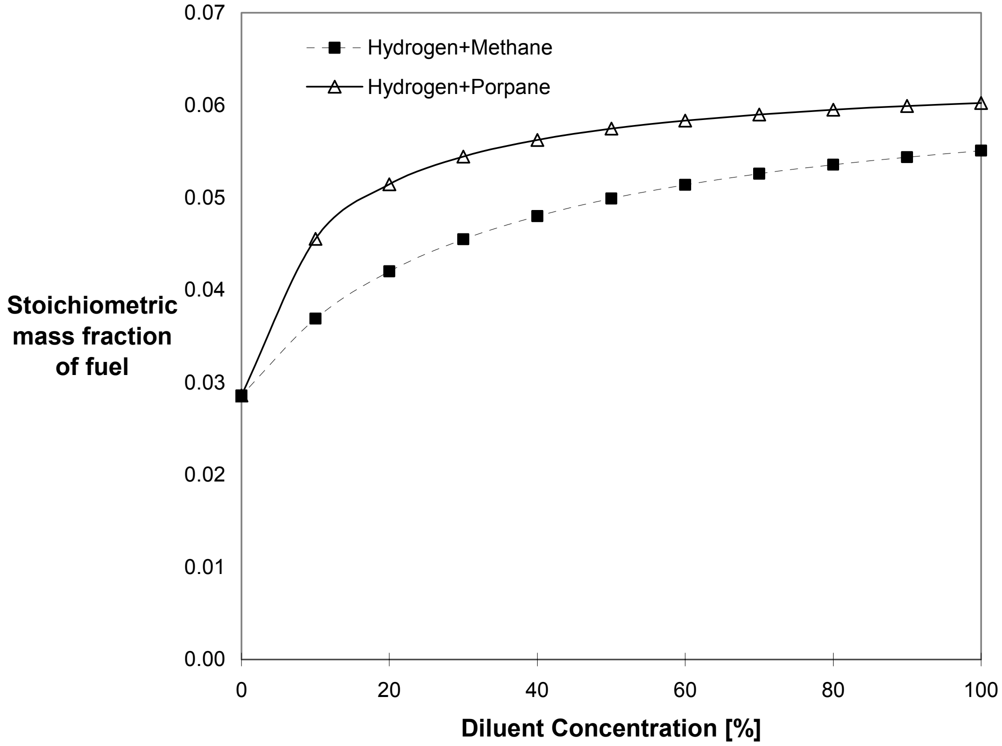

Figure 7 Comparison of stoichiometric mass fraction of fuel of the H_2-C_3H_8 mixtures and H_2-CH_4 mixtures as a function of diluent concentration.

5. CONCLUSIONS

- The flame lift-off height of the pure H_2 jet diffusion flame was found to increase with the jet velocity. Measured H_2 lift-off heights agreed well with other published data of Kalghatgi (1981) and of Cheng and Chiou (1998). Agreement with previously published correlations was found to depend on the value used for the maximum laminar burning velocity of H_2. A better fit between measurements and predictions was found if the stoichiometric burning velocity of H_2 is used. This raised an important issue as to whether the lifted flame base is located at the stoichiometric contour or at the location of maximum S_L.
- Comparisons of the stability of H_2-CO_2, H_2-C_3H_8 and H_2-CH_4 flames showed that the addition of C_3H_8 on hydrogen flames required least liftoff velocity and produced highest liftoff height among three additive gases. C_3H_8 is most effective in producing lifted flames.
- The experimental results also showed that C_3H_8 addition is more effective in blow-out hydrogen flame than CO_2 addition. CO_2 addition is more effective in blow-off attached hydrogen/additives flames than CH_4 addition.

- The effects of methane on the hydrogen flame were different from that for propane and had similarities to the ones of carbon dioxide. At high concentration, direct blow-off of the methane/hydrogen was observed.
- The lift-off process of H_2-C_3H_8 , H_2-CH_4 and H_2-CO_2 jet flames was controlled more by the chemical kinetics than by isothermal mixing processes. The addition of C_3H_8 acted as a sink for the active radicals that are of importance in the combustion chemistry of H_2. The addition of C_3H_8 also detrimentally affected the laminar burning velocity of H_2-C_3H_8 mixtures, causing higher lift-off heights for these mixtures. The CO_2 on the other hand, acted as an inert and therefore did not have a major effect on the H_2 reaction chain. Thus CO_2 did not significantly affect the flame stability. This was further confirmed by the use of Ar, which affected the flame stability in the same way as that observed with CO_2.

- Addition of C_3H_8 and CH_4 also changes the mixture's stoichiometric fuel/ air ratio. The H_2-C_3H_8 flame's stoichiometric fuel mass fraction increases with C_3H_8 concentration, and therefore the H_2-C_3H_8 flame requires a longer distance to attain stoichiometric concentration and higher exit velocity to cause flame blow out than for H_2-CO_2 mixtures.

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