ASSESSMENT OF THE EFFECTS OF INERT GAS AND HYDROCARBON FUEL DILUTION ON HYDROGEN FLAMES

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Abstract

To advance hydrogen into the energy market, it is necessary to consider risk assessment for scenarios that are complicated by accidental hydrogen release mixing with other combustible hydrocarbon fuels. The paper is aimed at examining the effect of mixing the hydrocarbon and inert gas into the hydrogen flame on the kinetic mechanisms, the laminar burning velocity and the flame stability. The influences of hydrogen concentration on the flame burning velocity were determined for the hydrogen/propane $(H_2-C_3H_8)$, hydrogen/ethane $(H_2-C_2H_6)$, hydrogen/methane (H_2-CH_4) , and hydrogen/carbon dioxide (H_2-CO_2) mixtures. Experimental tests were carried out to determine the lift-off, blow-out and blow-off stability limits of H_2 , $H_2-C_3H_8$, $H_2-C_2H_6$, H_2-CH_4 and H_2-CO_2 jet flames in a 2 mm diameter burner. The kinetic mechanisms of hydrogen interacting with C_3 , C_2 and C_1 fuels is analysed using the kinetic mechanisms for hydrocarbon combustion.

Keywords: hydrogen, hydrogen/hydrocarbon fuel, chemical kinetics, stability limits,

1.0 INTRODUCTION

The kinetic mechanisms of premixed hydrogen flames has been studied extensively in the past a few decades [1-4]. The full chemical kinetic mechanisms for hydrogen flame has been well established and involve eight reacting species (H₂, O₂, H, O, OH, HO₂, H₂O₂ and H₂O). Comparing to hydrogen flames, the kinetic mechanisms for hydrocarbon combustion are much more complicated. For oxidation of hydrocarbons up to propane, the order of hundred reactions around 30 species were found for a sufficiently accurate calculations of the chemistry [4]. In the recent development of numerical studies of flame structures, it was found necessary to develop reduced mechanisms for simplified descriptions and reduction of the chemistry to the smallest number of global steps. It is expected that the kinetic mechanisms would be even more complicated if the hydrogen is mixed with hydrocarbon fuel. So far there is very little kinetic information available on hydrogen-hydrocarbon mixtures. To carry out risk assessment for hydrogen energy system, the scenarios of the accidental hydrogen release mixing with other combustible hydrocarbon fuels have to be assessed. Therefore the kinetic mechanisms for hydrogen and hydrocarbon have to be established to allow detailed numerical simulation of flame structure and the hazard assessment. This paper examines the lift-off and blow-out stability of the hydrogen/hydrocarbon flames. The flame lift-off and blow-out parameters are strongly influenced by the flame burning velocity, which are directly linked to the kinetic mechanisms in the combustion reactions. The paper is aimed at examining the effect of mixing the hydrocarbon and inert gas into the hydrogen flame on the kinetic mechanisms, the laminar burning velocity and the flame stability.

2.0 EXPERIMENTAL TESTS ON THE FLAME STABILITY

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 channelling 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 in 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 of hydrogen, hydrogen/carbon dioxide flames. All flame images were captured using digital camera and processed using computer graphic packages.

2.1 Lift-off Velocity

A comparison of effect of propane addition and methane additions on lift-off velocity is shown in Figure 1. It is shown that for the same volume concentration, hydrogen/methane flames require much higher lift-off velocity than hydrogen/propane flames.



Figure 1: Comparison of the effect of C₃H₈, CO₂ and CH₄ addition on the flame lift-off velocity.

2.2 Blow-out and Blow-off Velocity

The experimental measured blow-out or blow-off velocities for hydrogen/propane (H_2 - C_3H_8), hydrogen/methane (H_2 - CH_4), and hydrogen/carbon dioxide (H_2 - CO_2) flames are presented in Figure 2. The comparison shows that for the same additive concentration, H_2 - CH_4 flames exhibit higher blow-out or blow-off velocity than H_2 - C_3H_8 or H_2 - CO_2 flames. However the blow-out or blow-off velocities are in the same order for H_2 - C_3H_8 or H_2 - CO_2 flames.

Based on the Premixed Combustion Model assumption that the incoming gas velocity at the stabilization zone balances the local turbulent velocity S_T , Kalghatgi [5] successfully derived an empirical formula that correlates the blow-out velocity, $U_{blowout}$, for various fuel mixtures (CH₄/Air, CH₄/CO₂, C₃H₈/Air and C₃H₈/CO₂) and burner diameters. The correlation for blow-out velocity is expressed as:

$$U_{blowout} = S_{L,\max} \left(\frac{\rho_{\infty}}{\rho_{Jet}}\right)^{1.5} \times 0.017 \operatorname{Re}_{H} \left(1 - 3.5 \times 10^{-6} \operatorname{Re}_{H}\right)$$
(1)

Where Re_{H} is Reynolds number based on dimensionless height, H, which is obtained from the following formula:

$$H = \left\{ 4 \frac{Y_o}{Y_{ST}} \left(\frac{\rho_{Jet}}{\rho_{\infty}} \right)^{0.5} + 5.8 \right\} d_o$$
(2)

Where γ_0 is the fuel mass fraction at burner exit; γ_{ST} is the stoichiometric mass fraction.

Since the blow-out velocity is proportion to the fuel maximum laminar burning velocity, the flame blow-out stability data in Figure 2 indicated that for the same concentration, H_2/CH_4 is more active than H_2/C_3H_8 , H_2/CO_2 mixtures, which is in the contrary to pure CH_4 and C_3H_8 fuels where C_3H_8 is more active than CH_4 . The presence of hydrogen has reversed the reactivity order of the C_1 and C_3 fuels.



Figure 2: Comparison of the effect of C_3H_8 , CO_2 and CH_4 addition on the flame blow-out or blow-off velocity.

3.0 LAMINAR BURNING VELOCITY

Experimental studies of the laminar or fundamental flame velocity of hydrogen-air mixtures, hydrogen/hydrocarbon mixtures and hydrogen rich syngas have been reported in a number of studies [6-12] and selected burning velocity values are presented in Figure 3. The measured laminar burning velocity of pure hydrogen flames peaked at a rich mixture of equivalence ratio 1.8. However the hydrogen/hydrocarbon mixtures assembled the features of hydrocarbon fuels, with the peak close to the stoichiometric. Comparing the magnitude of the burning velocity for the same equivalent ratio, the descending order of the burning velocity for pure hydrocarbon fuels is C_3H_8 , C_2H_6 and CH_4 . Comparing the hydrogen-hydrocarbon flames, the descending order is H_2 - C_1H_6 and H_2 - C_3H_8 . It is confirmed that the reactive order for C_3H_8 , C_2H_6 and CH_4 fuels seems reversed if hydrogen is added. This is more clearly shown in Figure 4 if the values of maximum laminar burning velocity is compared for H_2 - C_3H_8 , H_2 - C_2H_6 and H_2 - CH_4 mixtures. This clearly indicates that the role of hydrogen in the chemical kinetic mechanisms for hydrogen/hydrocarbon mixtures needs to be examined.



Figure 3: The laminar burning velocity of H₂, H₂-C₃H₈, H₂-C₂H₆ and H₂-CH₄ mixtures.



Figure 4: The ratio of maximum burning velocity to hydrogen maximum laminar burning velocity for H_2 - C_3H_8 , H_2 - C_2H_6 and H_2 - CH_4 mixtures.

4.0 ANALYSIS OF KINETIC MECHANISMS

At present, there is no established detailed or reduced kinetic pathway for hydrogen/hydrocarbon mixtures. In this paper, the analysis is based on comparing the kinetic mechanism established for pure hydrogen premixed flames and the kinetic mechanism for hydrocarbon fuels up to propane. A detailed kinetic mechanism by Warnatz [2, 4] for hydrogen and hydrocarbon fuels up to propane is listed in Table 1.

The full 8-species kinetic mechanism for premixed hydrogen flame is presented in the reactions 1 to 15 in the Table 1. For the kinetic mechanism of C_3 -chain, the dominant consumption path of C_3H_8 is to form normal- and iso- C_3H_7 radicals via reactions 77 & 78 with radical H, reaction 79 & 80 with O and reactions 81 & 82 with OH. Both n- C_3H_7 and i- C_3H_7 radicals decompose quickly to form C_2H_4 and CH₃ via reaction 87 and with minor formation of C_3H_6 via reaction 88 & 89. The remaining reaction paths produce subsequent consumes of C_3 , C_2 and C chains involve with attacks of free radicals H and OH detailed in the Table 1. Ethane (C₂H₆) combustion is initiated with H, OH and O radicals in reactions 44 to 46. The path way for Methane (CH₄) is presented in reactions 18 to 40.

The analysis the reactions shows that the C_3 , C_2 and C chains use radicals H, OH and O to break the C-H bond and C-C bond and to decompose to lighter hydrocarbons during combustion. Comparing hydrogen flames and hydrocarbon flames, hydrogen flames have high concentration of H, OH and O radicals, which are responsible for the high burning velocity, in the contrary the hydrocarbon flames have low concentration of H, OH and O radicals.

When hydrogen is mixed with hydrocarbon, hydrogen has provided a pool of H, OH and O radicals. However the reaction pathway is dominated by hydrocarbon consumptions, the radicals are absorbed by the decomposition of carbon chains and therefore the flames actually exhibit the features of hydrocarbon flames unless very large proportion of hydrogen (over 50% hydrogen) is present. The demand for H, OH and O radicals increase with the carbon numbers, the heavier hydrocarbon such as C_3H_8 would absorb much more radicals than the lighter hydrocarbon such as CH_4 . Therefore the improvement on burning velocity by hydrogen is less effective in the carbon heavier fuel due to the disappearance of the radicals. This results that the H₂-CH₄ has slight higher burning velocity than H₂- C_3H_8 . For the hydrogen rich syngas and H₂-CO₂, the reaction pathways are clearly dominated by hydrogen reactions and produce higher burning velocity than hydrogen/hydrocarbon.

No.	Reaction	А	n	Е				
		[cm,mol,s]		kJ/mole				
	Mechanism of the oxidation of H ₂ and CO							
H ₂ -O ₂ Chain propagation and branching reactions								
1	$O_2 + H \rightarrow OH + O$	1.200E+17	-0.91	70.30				
2	$OH + O \rightarrow O_2 + H$	7.100E+15	-0.91	0.00				
3	$H_2 + O \rightarrow OH + H$	1.500E+07	2.00	31.60				
4	$OH + H \rightarrow H_2 + O$	6.700E+06	2.00	23.30				
5	$H_2 + OH \rightarrow H_2O + H$	1.000E+08	1.60	13.80				
6	$H_2O + H \rightarrow H_2 + OH$	4.600E+08	1.60	77.70				
7	$OH + OH \rightarrow H_2O + O$	1.500E+09	1.14	0.00				
8	$H_2O + O \rightarrow OH$	1.500E+10	1.14	72.20				
Recombination reactions								
9	$\mathrm{H} + \mathrm{H} + \mathrm{M}' \rightarrow \mathrm{H}_2 + \mathrm{M}'$	1.800E+18	-1.00	0.00				
10	$OH + H + M' \rightarrow H_2O + M'$	2.200E+22	-2.00	0.00				

Table 1. Warnatz	(1985)) [5]	l kinetic	mechanisms	for h	vdrocarbon	combustion
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HO ₂ Formation and Consumption							
11	$O_2 + H + M' \rightarrow HO_2 + M'$	2.000E+18	-0.80	0.00			
12	$\mathrm{HO}_2 + \mathrm{H} \rightarrow \mathrm{OH} + \mathrm{OH}$	1.500E+14	0.00	4.20			
13	$HO_2 + H \rightarrow H_2 + O_2$	2.500E+13	0.00	2.90			
14	$HO_2 + O \rightarrow OH + O_2$	2.000E+13	0.00	0.00			
15	$HO_2 + OH \rightarrow H_2O + O_2$	2.000E+13	0.00	0.00			
CO Oxid	ation						
16	$CO + OH \rightarrow CO_2 + H$	4.400E+06	1.50	-3.10			
17	$\rm CO_2 + H \rightarrow \rm CO + OH$	1.600E+14	0.00	110.00			
	Mechanism of the oxid	lation of C_1/C_2 hydrocarb	oons				
Formatio	n and consumption of CH_4						
18	$CH_3 + H \rightarrow CH_4$	6.000E+16	-1.00	0.00			
19	$CH_4 + H \rightarrow CH_3 + H_2$	2.200E+04	3.00	36.60			
20	$CH_3 + H_2 \rightarrow CH_4 + H$	6.600E+02	3.00	32.40			
21	$CH_4 + O \rightarrow CH_3 + OH$	1.200E+07	2.10	31.90			
22	$CH_3 + OH \rightarrow CH_4 + O$	1.300E+05	2.10	19.60			
23	$CH_4 + OH \rightarrow CH_3 + H_2O$	1.600E+06	2.10	10.30			
24	$CH_3 + H_2O \rightarrow CH_4 + OH$	2.900E+05	2.10	70.30			
CH ₃ Consumption							
25	$CH_3 + O \rightarrow CH_2O + H$	7.000E+13	0.00	0.00			
CH ₂ O Co	nsumption						
26	$CH_2O + H \rightarrow CHO + H_2$	2.500E+13	0.00	16.70			
27	$CH_2O + O \rightarrow CHO + OH$	3.500E+13	0.00	14.70			
28	$CH_2O + OH \rightarrow CHO + H_2O$	3.000E+13	0.00	5.00			
CHO Cor	isumption						
29	$\rm CHO + \rm H \rightarrow \rm CO + \rm H_2$	2.000E+14	0.00	0.00			
30	$CHO + O \rightarrow CO + OH$	3.000E+13	0.00	0.00			
31	$\rm CHO + O \rightarrow \rm CO_2 + \rm H$	3.000E+13	0.00	0.00			
32	$\rm CHO + OH \rightarrow \rm CO + \rm H_2O$	5.000E+12	0.00	0.00			
33	$\rm CHO + O_2 \rightarrow \rm CO + \rm HO_2$	3.000E+12	0.00	0.00			
34	$\mathrm{CHO} + \mathrm{M}' \rightarrow \mathrm{CO} + \mathrm{H} + \mathrm{M}'$	7.100E+14	0.00	70.30			
CH ₂ Consumption							
35	$CH_2 + H \rightarrow CH + H_2$	4.000E+13	0.00	0.00			
36	$CH_2 + O \rightarrow CO + H + H$	5.000E+13	0.00	0.00			
37	$CH_2 + O_2 \rightarrow CO_2 + H + H$	1.300E+13	0.00	6.30			
38	$CH_2 + CH_3 \rightarrow C_2H_4 + H$	4.000E+13	0.00	0.00			
CH Cons	umption						
39	$CH + O \rightarrow CO + H$	4.000E+13	0.00	0.00			
40	$CH + O_2 \rightarrow CO + OH$	2.000E+13	0.00	0.00			
Formation of C ₂ hydrocarbons by CH ₃ recombination							
41	$CH_3 + CH_3 \rightarrow C_2H_6$	2.400E+14	-0.40	0.00			
42	$CH_3 + CH_3 \rightarrow C_2H_5 + H$	8.000E+13	0.00	111.00			
43	$CH_3 + CH_3 \rightarrow C_2H_4 + H$	1.000E+16	0.00	134.00			
C ₂ H ₆ Consumption							

44	$C_2H_6 + H \rightarrow C_2H_5 + H_2$	5.400E+02	3.50	21.80		
45	$C_2H_6 + O \rightarrow C_2H_5 + OH$	3.000E+07	2.00	21.40		
46	$C_2H_6 + OH \rightarrow C_2H_5 + H_2O$	6.300E+06	2.00	2.70		
47	$C_2H_5 + H \rightarrow CH_3 + CH_3$	3.000E+13	0.00	0.00		
48	$C_2H_5 + H \rightarrow CH_3 + CH_3$	5.000E+13	0.00	0.00		
49	$C_2H_5 + O_2 \rightarrow C_2H_4 + HO_2$	2.000E+13	0.00	20.90		
50	$C_2H_5 \rightarrow CH_4 + H$	2.000E+13	0.00	166.00		
C ₂ H ₄ Cor	nsumption					
51	$C_2H_4 + H \rightarrow C_2H_5$	1.000E+13	0.00	6.30		
52	$C_2H_4 + O \rightarrow CHO + CH_3$	1.600E+09	1.20	3.10		
53	$C_2H_4 + OH \rightarrow C_2H_3 + H_2O$	7.000E+13	0.00	12.60		
54	$C_2H_4 + H \rightarrow C_2H_3 + H_2$	1.500E+14	0.00	42.70		
C ₂ H ₃ Cor	nsumption					
55	$C_2H_3 + H \rightarrow C_2H_2 + H_2$	2.000E+13	0.00	0.00		
56	$C_2H_3 + O_2 \rightarrow C_2H_2 + HO_2$	1.000E+12	0.00	0.00		
57	$C_2H_3 \rightarrow C_2H_2 + H$	1.000E+15	0.00	178.00		
C ₂ H ₂ Cor	isumption					
58	$C_2H_2 + H \rightarrow C_2H_3$	5.500E+12	0.00	10.10		
59	$C_2H_2 + O \rightarrow CH_2 + CO$	4.100E+08	1.50	7.10		
60	$C_2H_2 + OH \rightarrow C_2HCO + H$	3.000E+12	0.00	4.60		
CH ₃ CHO	Consumption					
61	$CH_3CHO + H \rightarrow CH_3 + CO + H$	4.000E+13	0.00	17.60		
62	$CH_3CHO + O \rightarrow CH_3 + CO + OH$	5.000E+12	0.00	7.50		
63	$CH_3CHO + OH \rightarrow CH_3 + CO + H_2O$	1.000E+13	0.00	0.00		
CH ₂ CO C	Consumption					
64	$CH_2CO + H \rightarrow CH_3 + CO$	7.000E+12	0.00	12.60		
65	$CH_2CO + O \rightarrow CHO + CHO$	2.000E+13	0.00	9.60		
66	$CH_2CO + OH \rightarrow CH_2 + CHO$	1.000E+13	0.00	0.00		
67	$\rm CH_2\rm CO + \rm M \rightarrow \rm CH_2 + \rm CO + \rm M$	1.000E+16	0.00	248.00		
CHCO F	ormation and consumption					
68	$C_2H_2 + OH \rightarrow CHCO + H$	4.300E+14	0.00	50.70		
69	$CHCO + H \rightarrow CH_2 + CO$	3.000E+13	0.00	0.00		
70	$\rm CHCO + O \rightarrow \rm CO + \rm CO + \rm H$	1.200E+12	0.00	0.00		
C ₂ H Formation and consumption						
71	$C_2H_2 + H \rightarrow C_2H + H_2$	1.500E+14	0.00	79.60		
72	$\mathrm{C_2H_2} + \mathrm{OH} \rightarrow \mathrm{C_2H} + \mathrm{H_2O}$	1.000E+13	0.00	29.30		
73	$C_2H + O \rightarrow CO + CH$	1.000E+13	0.00	0.00		
74	$C_2H + H_2 \rightarrow C_2H_2 + H$	3.500E+12	0.00	8.80		
75	$C_2H + O_2 \rightarrow CO + CHO$	5.000E+13	0.00	6.30		
Mechanism of the oxidation of C ₃ hydrocarbons (C ₃ H ₄ chemistry excluded)						
C ₃ H ₈ Formation and consumption						
76	$C_2H_5 + CH_3 \rightarrow C_3H_8$	7.000E+12	0.00	0.00		
77	$\mathrm{C}_{3}\mathrm{H}_{8} + \mathrm{H} \rightarrow \mathrm{n}\text{-}\mathrm{C}_{3}\mathrm{H}_{7} + \mathrm{H}_{2}$	1.300E+14	0.00	40.60		
78	$C_3H_8 + H \rightarrow i-C_3H_7 + H_2$	1.000E+14	0.00	34.90		

79	$\mathrm{C_3H_8} + \mathrm{O} \rightarrow \mathrm{n}\text{-}\mathrm{C_3H_7} + \mathrm{OH}$	3.000E+13	0.00	24.10			
80	$C_3H_8 + O \rightarrow i-C_3H_7 + OH$	2.600E+13	0.00	18.7			
81	$\mathrm{C_3H_8} + \mathrm{OH} \rightarrow \mathrm{n}\text{-}\mathrm{C_3H_7} + \mathrm{H_2O}$	3.700E+12	0.00	6.90			
82	$\mathrm{C_3H_8} + \mathrm{OH} \rightarrow \mathrm{i\text{-}C_3H_7} + \mathrm{H_2O}$	2.800E+12	0.00	3.60			
C ₃ H ₇ Con	sumption						
83	$n-C_3H_7 + H \rightarrow C_3H_8$	2.000E+13	0.00	0.00			
84	$i-C_3H_7 + H \rightarrow C_3H_8$	2.000E+13	0.00	0.00			
85	$\text{n-}C_3\text{H}_7 + \text{O}_2 \rightarrow \text{C}_3\text{H}_6 + \text{HO}_2$	1.000E+12	0.00	20.90			
86	$i\text{-}C_3\text{H}_7 + \text{O}_2 \rightarrow \text{C}_3\text{H}_6 + \text{HO}_2$	1.000E+12	0.00	12.50			
87	$n\text{-}C_3H_7 \rightarrow C_2H_4 + CH_3$	3.000E+14	0.00	138.00			
88	$n\text{-}C_3H_7 \rightarrow C_3H_6 + H$	1.000E+14	0.00	156.10			
89	i - $C_3H_7 \rightarrow C_3H_6$ + H	2.000E+14	0.00	161.90			
C ₃ H ₆ Consumption							
90	$n-C_3H_6 + H \rightarrow n-C_3H_7$	4.000E+12	0.00	11.00			
91	$\mathrm{i\text{-}}\mathrm{C}_3\mathrm{H}_6 + \mathrm{H} \rightarrow \mathrm{i\text{-}}\mathrm{C}_3\mathrm{H}_7$	4.000E+13	0.00	4.00			
92	$\mathrm{C_3H_6} + \mathrm{O} \rightarrow \mathrm{CH_3} + \mathrm{CH_3} + \mathrm{CO}$	5.000E+12	0.00	1.90			
93	$C_3H_6 + OH \rightarrow CH_3CHO + CH_3$	1.000E+13	0.00	0.00			

5.0 CONCLUSIONS

- Comparisons of the stability of H₂-CO₂, H₂-C₃H₈ and H₂-CH₄ flames shows that descending order of blow-out or blow-off velocity is H₂-CH₄, H₂-O₂ and H₂- C₃H₈. The lift-off and blow-out process of H₂-C₃H₈, H₂-CH₄ and H₂-CO₂ jet flames was strongly influenced by the chemical kinetics. Analysis using the stability theory indicates that reactive order of the flames is H₂-CH₄, H₂-O₂ and H₂- C₃H₈ flames.
- Comparing the magnitude of the burning velocity of pure C₃H₈, C₂H₆ and CH₄ fuels and H₂-CH₄, H₂-C₂H₆ and H₂- C₃H₈ mixtures confirms that the reactive order for C₃, C₂ and C₁ fuels seems reversed if hydrogen is added. This findings supports the conclusions made in the flame lift-off and blow-out stability study.
- The analysis on the kinetic pathways indicates that hydrocarbons act as a sink for the active radicals that are of importance in the combustion chemistry of H₂. The hydrocarbon is the dominant element in determine the burning velocity of hydrogen/hydrocarbon mixtures. In H₂-CO₂ mixture, hydrogen reactions remand as the main kinetic mechanism.
- Based on the kinetic mechanism analysis, it is possible to produce reduced kinetic mechanisms for hydrogen/hydrocarbon, which would be important for numerical simulations of the flame structure, propagation and hazard analysis.

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