BIOGAS COMBUSTION CHARACTERISTICS UNDER VARYING CARBON DIOXIDE DILUTION AND HYDROGEN ENRICHMENT

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ABSTRACT

This study investigates the combustion characteristics of four different types of gaseous fuels namely methane (CH₄), biogas, simulated biogas with varying carbon dioxide (CO₂) content and hydrogen enriched biogas under atmospheric condition. Flammability, laminar burning velocity and flame stability of each fuel are among the combustion characteristics investigated using spherical flame method. Measurement of these parameters is important to explain the effects of CO_2 and hydrogen on biogas combustion which are still lacking in literature. CH₄ flammability range was found to be within the equivalence ratio of 0.7 to 1.3 with peak laminar burning velocity at approximately 36 cm/s which agrees well with previous findings. For biogas, flammability range narrows to equivalence ratio range of 0.6 to 0.9 with a peak laminar burning velocity of around 24 cm/s. For simulated biogas, as CO₂ content increased, the flammability range tended to become narrower with appreciable decrease in laminar burning velocity. Peak laminar burning velocity value steadily decreased to 21%, 34% and 45% as CO2 content was increased from 20% to 40% and 50% respectively. CO₂ could slow down the reactions that produce radicals important for CH₄ dissociation. It could also modify mass and thermal diffusion pattern as indicated by the corresponding changes in Markstein length. For hydrogen enriched biogas, the flammability limits widened to the leaner side from equivalence ratio of 0.4 to 0.9 for 30% and 40% enrichment. Both flame speed and laminar burning velocity were enhanced with hydrogen enrichment especially at 30% and 40% which led to significant increase in maximum laminar burning velocity to 52 % and 88 % respectively. Flame appeared to become less stable under leaner conditions as supported by the occurrence of buoyancy and mild cellularity at the equivalence ratio of 0.4 and 0.5 under 30% and 40% hydrogen enrichment. Simulation revealed dramatic increase in H radical at 30% hydrogen enrichment onwards. These observations imply the significance of hydrogen on biogas combustion both on laminar burning velocity and flame stability.

ABSTRAK

Kajian ini bertujuan menyelidik sifat-sifat pembakaran bagi empat jenis bahanapi gas iaitu metana (CH₄), biogas, biogas tersimulasi dengan variasi kandungan karbon dioksida (CO₂) dan biogas yang diperkaya dengan hidrogen di bawah keadaan atmosfera. Kebolehbakaran, halaju pembakaran laminar dan kestabilan nyala adalah antara sifat-sifat pembakaran yang dikaji menggunakan kaedah nyala sfera. Pengukuran parameter-parameter ini adalah penting bagi menjelaskan kesan-kesan CO₂ dan hidrogen terhadap pembakaran biogas yang mana masih kurang dilaporkan. Julat kebolehbakaran CH₄ ialah diantara nisbah kesetaraan 0.7 hingga 1.3 dengan puncak halaju pembakaran laminar kira-kira 36 cm/s yang mana menyamai hasil kajian terdahulu. Bagi biogas, julat kebolehbakaran mengecil antara nisbah kesetaraan 0.6 hingga 0.9 dengan nilai puncak halaju pembakaran laminar pada kira-kira 24 cm/s. Bagi biogas tersimulasi pula apabila kandungan CO₂ meningkat, julat kebolehbakaran didapati mengecil dengan halaju pembakaran laminar menurun. Nilai puncak halaju pembakaran laminar berkurang sebanyak 21%, 34% dan 45% apabila kandungan CO₂ bertambah masing-masing dari 20% kepada 40% dan 50%. CO₂ berupaya memperlahankan tindakbalas-tindakbalas yang menghasilkan radikal-radikal penting dalam penguraian CH₄. Ia juga berupaya mengubah corak penyebaran haba dan jisim seperti ditunjukkan oleh perubahan dalam kepanjangan Markstein. Bagi biogas yang diperkaya hidrogen, julat kebolehbakaran bagi kandungan hidrogen 30% dan 40% didapati melebar dari nisbah kesetaraan 0.4 hingga 0.9. Kedua-dua laju nyala dan halaju pembakaran laminar telah ditingkatkan dengan pengayaan hidrogen khususnya pada 30% dan 40% yang mana telah meningkatkan halaju pembakaran laminar maksimum secara ketara masing-masing kepada 52% dan 88%. Nyala kelihatan kurang stabil dibawah keadaan yang lebih langsing disokong dengan nyalaan apungan dan selulariti sederhana pada nisbah kesetaraan 0.4 dan 0.5 di bawah pengayaan hidrogen 30% dan 40%. Simulasi mendedahkan peningkatan radikal H yang dramatik bermula dari kandungan hidrogen 30%. Ini menunjukkan pengaruh hidrogen yang amat ketara dalam pembakaran biogas dari segi halaju dan juga kestabilan nyala.

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LIST OF ABBREVIATIONS

BG50	Biogas with 50% methane
BG60	Biogas with 60% methane
BG80	Biogas with 80% methane
BOD	Biochemical oxygen demand
BTE	Brake thermal efficiency
CH ₄	Methane
CJ	Chapman Jouget
СО	Carbon Monoxide
CO_2	Carbon Dioxide
COD	Chemical oxygen demand
СРО	Crude palm oil
CVCC	Constant volume combustion chamber
DRM	Developed Reduced Mechanism
GHG	Green house gasses
GRI	Gas Research Institute
ICE	Internal combustion engine
H ₂	Hydrogen
LPG	Liquefied petroleum gas
NOx	Nitrogen oxides
NUIG	National University of Ireland Galway
РАН	Polycyclic Aromatic Hydrocarbon
POME	Palm oil mill effluent
SI	Spark ignition
STP	Standard temperature and pressure
TCD	Thermal conductivity detector
UTM	Universiti Teknologi Malaysia

LIST OF SYMBOLS

α	Flame stretch		
A	Pre-exponenial Factor		
A_f	Spherical Flame Surface Area		
<i>a</i> _{tt}	Tangential Strain Rate		
β	Secondary Temperature Dependency		
С	Capacitance		
χ	Hydrogen enrichment percentage		
$\Delta_r H^{\circ}$	Enthalpy of combustion		
E	Energy		
$E_{l,min}$	Minimum ignition energy		
d_q	Quenching distance		
δ_l	Laminar flame thickness		
δ_D	Flame thickness		
D_u	Mass diffusivity		
f	Mass flux		
Ka	Karlovitz Number		
k	Rate Constant		
$\lambda_{\rm v}$	Air/Fuel Ratio		
λ	Thermal conductivity		
m_{LZ}	Mass of Air		
m_B	Maximum Convertible Fuel Mass		
L_b	Markstein Length		
Le	Lewis Number		
L_{min}	Fuel Specific Stoichiometric Air Requirement		
$(A/F)_{stoic}$	Stoichiometric Air-Fuel Ratio		
Μ	Mach Number		
Ma	Markstein Number		
MW _{air}	Molecular Weight of Air		
MW_{fuel}	Molecular Weight of Fuel		
$ ho_b$	Burned Gas Density		

$ ho_u$	Unburned Gas Density
ϕ	Equivalence Ratio
R	Universal Gas Constant
\mathbb{R}^2	Coefficient of Determination
Rec	Critical Reynolds Number
R_{f}	Flame Radius
$R_{f,c}$	Critical Radius at the Onset of Cellularity
γ	Specific Heat Ratio
σ	Density ratio
S_n	Stretched Flame Speed
S_s	Unstretched Flame Speed
u_L	Laminar Burning Velocity

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Depleting supply and stricter emission regulation has motivated the search for alternative fuels that are both green and sustainable to meet the increasing global demand for energy and emission regulation (Basha *et al.*, 2009; Hosseini and Wahid, 2013). Fossil fuel is still the main energy source for transportation, industrial and agricultural activities. Based on current consumption trend, it is projected that fossil fuel will become fully exhausted by 2050 (Basha *et al.*, 2009).

Biogas could be one of the potential alternative fuels to cater future energy demand. It is a product of anaerobic fermentation of various wastes could be the energy of the future. It is mainly consists of around 50-70% methane and 30-50% carbon dioxide with several other trace gases at smaller quantity. Abundant supply could be the main factor that could turn biogas as the promising fuel that could meet future energy demand. Biogas could be the potential fuel especially in the power plant to meet the ever increasing demand for energy. It has drawn the attention of researchers to the extent that many studies have been conducted to assess the potential of biogas to fuel both stationary and non-stationary engines (Razbani *et al.*, 2011). The use of biogas can also help in reducing Green House Gas (GHG) emission as biogas combustion produces lesser pollutants than fossil fuels (Schoen and Bagley, 2012).

At present, Germany is the global leader in biogas utilization with almost 600% growth in power generation from 60 kW_e in 1999 to 350 kW_e in 2008 (Poeschl *et al.*, 2010). Biogas utilization by means of combustion is mainly for generating electricity, powering vehicle, domestic use etc. (Poeschl *et al.*, 2010; Soleimani, 2010). Biogas has a big potential to be used in any type of engine provided the correct technique on its utilization is observed.

Combustion is one way to extract the energy contained within the chemical bonds of the fuel molecules. During combustion, fuel molecules are oxidized via a chemical reaction with oxygen releasing heat and forming certain combustion products (Moran and Shapiro, 2004). However, knowledge on the combustion characteristics of biogas is necessary to optimize the use of biogas while at the same time reducing the unwanted combustion by-products (Metgalchi and Keck, 1980). The interaction between its major components i.e. CH₄ and CO₂ could lead to unpredictable combustion characteristics that could hinder the commercial use of biogas (Fischer and Jiang, 2015). Thus, it is imperative to study biogas combustion characteristics at its fundamental level. Knowledge about its combustion characteristics could also be useful in the design of practical combustors that can efficiently run on biogas (Diaz-Gonzalez at al., 2009).

This study focuses on the laminar fully premixed biogas combustion. This kind of combustion permits the burning of the fuel at equivalence ratio other than the stoichiometric. Thus lower temperature combustion can be achieved under the lean condition. Laminar burning velocity, flammability limit, flame stability and Markstein length are among the properties widely used to describe the combustion behavior of certain fuel. These properties are unique for every different fuel as they are directly influenced by fuel properties to a certain extent.

It was well reported that the presence of CO_2 can significantly affect the combustion characteristics of biogas in certain way. CO_2 can act as a diluent absorbing some of the heat generated during combustion. This implies that the flame propagation, flammability limits and flame stability are all affected by the presence of CO_2 (Hinton and Stone, 2014). In addition, CO_2 can also promote flame buoyancy as it dilutes the concentration of the only flammable component of biogas which is methane per given biogas volume.

Laminar burning velocity is the velocity at which the combustible mixture propagates into the reaction zone. It directly influence the development of pressure especially within a closed system. It is also the most important flame property in spark ignited premixed combustion such as those found in automobiles' gasoline engine. The presence of CO_2 has been found to affect the laminar flame velocity of biogas where an increase in CO_2 concentration would cause a corresponding decrease in the laminar flame velocity. This reduction could become more apparent in richer fuel and oxidizer mixture.

Flammability limits which consist of the lower and upper limits define the range within which the fuel and oxidizer mixture is flammable. The lower and upper flammability limit are the leanest and richest mixture at which the fuel could combust with sustained flame respectively. If the lower flammability limit of biogas is lower than current gas such as natural or liquefied petroleum gas (LPG), biogas could combust with a much smaller volume compared to these gasses improving fuel economy. In fact, recent study by Anggono et al. (2014) reveals that the flammability limit of biogas depends on the initial pressure at which the biogas start to combust.

Markstein length is a parameter that describes the effects of changes in flame structure due to stretching to the flame speed (Moccia and D'Alesio, 2013). It is influenced by the chemical and transport properties of the reacting mixtures. It is important to evaluate this quantity as it could serve as an indicator whether the flame extinction could occur for a particular combustible fuel and oxidizer mixture.

Buoyancy is known to alter flame configuration distorting its structure and making analysis difficult. It is likely to occur for a lean mixture. The net convective motion resulting from buoyancy could affect combustion in certain way; it may hamper flame propagation and to some extent cause the flame to propagate back into the burnt region. Once this happens, flame will extinguish due to the low fuel and oxidizer concentration in that region.

As reported in literature, an increase in CO_2 concentration and reduces combustible mixture concentration approaching its lower flammability limit. In other words, the mixture becomes leaner in the presence of CO_2 . This could lead to various form of instabilities and to some extent flame extinction. Extinction would occur if there are stretching, nonequidiffusion (for the case where thermal diffusivity is higher than mass diffusivity) and incomplete reaction (for the case where mass diffusivity is higher than thermal diffusivity). The presence of CO_2 may contribute to flame extinction as it could absorb some of the heat generated during combustion.

1.2 Problem Statement

Palm Oil Mill Effluent (POME) is a type of effluent generated by palm oil processing. POME is usually channeled into treatment ponds where biogas is emitted during the treatment process. This gas could serve as an alternative energy as it contains significant amount of methane that could be harnessed to generate power especially in the form of electricity. As the second largest oil palm exporter, Malaysia should tap into this renewable resource in an attempt to implement sustainable energy generation that are both economical and environment friendly.

The benefits of tapping biogas are plentiful. First, it could reduce methane and CO_2 that would otherwise be released to the environment contributing to global warming. Anaerobic digester used in biogas capture instead of aerobic pond for POME treatment will minimize land area required for effluent treatment. Extra revenue can also be gained from palm oil milling process as it can also supply fuel.

However, the feasibility of using biogas as a fuel of power plant generator still remains subtle. The manner the combustion of biogas affects both the combustor in power plant generator and amount energy generated is of paramount importance. This would require a thorough study on biogas combustion characteristics.

The content of CO_2 may adversely affect biogas combustion as it would reduce calorific value, laminar burning velocity, flame speed and stability (Lee and Hwang, 2007). It was established in the literature that CO_2 may physically and chemically affect biogas combustion to a certain degree depending on its content. Physically, it could absorb some of the heat from combustion, reducing the temperature and also the flame speed. Chemically, it could reduce the formation of certain radicals important during ignition and subsequently the propagation of flame. There are also limited experimental studies in literature to elucidate such effects and data on are still scarce as there are limited experimental studies reported in literature (Zeng et al, 2018). Laminar burning velocity measurement is important to elucidate effects of CO_2 on biogas combustion. Numerical studies by Fischer and Jiang (2015) shows that CO_2 could significantly reduce biogas reactivity especially under higher CO_2 content (50%). This is supported by a substantial decrease in CO mass fraction that would affect certain reactions that are important to combustion. Therefore, a subsequent decrease in biogas laminar burning velocity is expected with an increase in CO_2 content. This would render the direct use of biogas is limited to certain types of combustors that can effectively accommodate biogas distinct and inferior combustion characteristics.

Introduction of certain additives to biogas may to some extent improve biogas combustion characteristics. Hydrogen is among the additives that has attracted the attention of researchers over the recent years. Due to its wider flammability and relatively high mass diffusivity compared to CH₄ and CO₂, hydrogen could improve biogas combustion especially in terms of flammability and laminar burning velocity. Studies on the combustion characteristics of hydrogen enriched biogas are still scarce in literature (Hu and Zhang, 2019). Cardona and Amell (2013) reported that the addition of hydrogen could enhance biogas laminar burning velocity. In terms of stability, the addition of hydrogen as additive has effectively widen the range within which the flame is stable (Leung and Wierzba, 2008). A recent study also reported an increase in flame stability via a small addition of hydrogen in biogas (Zhen et al., 2014). However this study only investigate biogas combustion under stoichiometric and rich due to the flammability limits of biogas. Biogas could only be ignited at φ of 0.9 with and addition of 10% (v/v) hydrogen. Extending the lean limit of biogas combustion could be both economically and environmentally favorable as less biogas would be burn and consequently producing less pollutants.

Understanding the possible combustion chemistry resulted from the use of hydrogen as an additive at certain percentage still remains as one of the most challenging task. To achieve this, it is imperative to study the effect of hydrogen on combustion characteristics such as flame propagation, flame stability and flammability limits. Quantitatively, this can be achieved by solving the governing equations together with detailed chemistry. This study therefore aimed at gaining insight of the underlying chemistry and physics involved that affect biogas combustion.

1.3 Research Objectives and Scopes

The objectives and scopes of this study could be summarized as:

- I. To investigate the combustion characteristics of simulated biogas with varying CO₂ content within equivalence ratio range of 0.6 to 1.1.
- II. To evaluate the combustion characteristics of actual biogas from POME with and without hydrogen as an additive within equivalence ratio range of 0.5 to 0.9.
- III. To modify reaction mechanisms of simulated and actual biogas combustion in the presence of hydrogen.

This study will only cover low quality biogas as it is the least studied biogas in literature. This study covers the study of combustion characteristics such as flammability limits, flame speed, laminar burning velocity, stretch rate, Markstein length and heat release rate of pure methane, biogas and biogas with hydrogen as additive. Percentage of CO2 and hydrogen was varied to investigate the effect of their variation on combustion characteristics. Pure methane combustion serves as the control experiment.

1.4 Significance of Study

Biogas could be considered as an untapped energy resource especially in Malaysia where much of the energy generated come from fossil fuel combustion. The use of biogas may reduce the country's dependence on the depleting fossil fuel that is has also saw dwindling price due to shortage in supply. Since biogas contains significant amount of CO_2 , it may also cause GHG emission problem if left untapped. The presence of CO_2 may also give biogas distinct combustion characteristics that are not always favorable. There are numerous data on laminar burning velocity of biogas both numerically and experimentally. Nonetheless, data on low quality biogas; biogas with 50% of CO_2 or larger by volume are still scarce. Although biogas generation has been utilized since the 1950's, and the principles of digestion are well documented, little is known about the burning of such gases. Most works in literature involve numerical simulation, with relatively fewer experimental data.

It is important to improve these shortcomings prior to the effective use of biogas as a fuel. This however, requires the understanding of the physics and chemistry involved during the combustion. Addition of hydrogen has been shown to favorably improve biogas combustion, but little is known about how hydrogen alters the chemistry and physics of biogas combustion. Observations that will be made through this study will hopefully contribute to the existing knowledge. This would provide an invaluable insight especially for the execution of further studies. The study of laminar premixed combustion of biogas may also serve as building blocks for turbulent combustion understanding. This could significantly help in the design of many practical combustors that mostly employ turbulent combustion.

Addition of hydrogen has been shown to favorably improve biogas combustion, but little is known about how hydrogen alters the chemistry and physics of biogas combustion (Hu and Zhang, 2019). Simulation with ANSYS Chemkin will help to determine important chemical reactions that affect biogas combustion. It would also allow the determination and modification of the rates of such reactions. Schlieren photography is a versatile and sophisticated technique that captures flame images due to density gradient. This implies that, even the slightest change in density resulting from combustion could be captured clearly as flame image and will help flame speed and stability analysis.

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ø	BG50	mole fraction	BG60	mole fraction	BG80	mole fraction
0.6	CH ₄	0.056				
	CO ₂	0.056				
	O ₂	0.187				
	N ₂	0.701				
0.7	CH ₄	0.064		0.066		0.067
	CO ₂	0.064		0.044		0.017
	O_2	0.183		0.187		0.134
	N_2	0.689		0.703		0.782
0.8	CH_4	0.072		0.074		0.076
	CO_2	0.072		0.049		0.019
	O_2	0.18		0.184		0.152
	N_2	0.676		0.693		0.753
0.9	CH ₄	0.079		0.082		0.085
	CO_2	0.079		0.054		0.021
	O_2	0.177		0.181		0.169
	N ₂	0.665		0.683		0.725
1	CH ₄	0.095		0.089		0.093
	CO_2	0.095		0.06		0.023
	O ₂	0.17		0.179		0.186
	N_2	0.64		0.672		0.698
		1				1
1.1	CH ₄					0.101
	CO_2					0.025
	O_2					0.202
	N_2					0.672

Appendix A Calculation of Fuel and Air Partial Pressure

Appendix B Apparatus



Figure B1 Ignition control box.



Figure B2 CVCC body dimension in millimeter.



Figure B3 CVCC with all the lines and instrumentation.

- 1. Pressure gauge
- 2. Electrode
- 3. Thermocouple 1
- 4. Thermocouple 2
- 5. Thermocouple 3
- 6. Mixer motor
- 7. Vacuum line
- 8. Fuel line
- 9. Exhaust line
- 10. Dry air line
- 11. Compressed air line
- 12. Collimating lens
- 13. Decollimating lens

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- 1. Flame propagation and burning rates of methane-air mixtures using schlieren photograph. (2016). Jurnal Teknologi 78/21-27.
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- The effects of CO₂ variation on hydrodynamic and thermal diffusive stability of biogas-air flame. (2018). Journal of Advanced Research in Fluid Mechanics and Thermal Sciences. Volume 52, Issue 2246 – 258.