CHARACTERIZATION OF FLAME PROPAGATION AND BURNING RATES OF VARIOUS MIXTURES OF BIOGAS AND NITROUS OXIDE

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DEDICATION

This work is completely dedicated to my respectful parents, my dear wife, and beloved children without whose constant support this thesis was not possible. They always inspire me. At the same time, my thanks also go to my caring friend Feras who supported me during this study.

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ABSTRACT

Biogas is a potential alternative energy source with low environmental impact. However, the practical applications of biogas are relatively limited due to the existence of carbon dioxide (CO_2) which acts as a diluent that decreases the calorific value and the burning rate of biogas. Nitrous oxide (N_2O) is known to be a powerful oxidizing agent for propulsion applications which can enhance the combustion rate of biogas, however, there is a lack of studies that investigate the fundamental characteristics of biogas-N_2O combustion. The aim of this study is to gain insight into the fundamental combustion characteristics of biogas-N_2O mixtures in terms of laminar burning velocity and flame stability. In the present work, spherically expanding flames following central ignition at constant volume combustion chamber (CVCC) were employed to investigate laminar burning velocity (LBV), hydrodynamic instability, and diffusive-thermal instability of biogas-N_2O mixtures at wide equivalence ratio;ϕ, from 0.6 to 1.4, at 303K and atmospheric pressure. Two mechanisms were used in CHEMKIN-PRO 17 software in order to estimate the predicted combustion characteristics of biogas-N_2O mixtures. The results indicate that the decline in LBVs was prominent in the fuel-rich mixtures than in the fuel-lean mixtures with CO_2 dilution. It is found that the influence of curvature on the flame front is weakened at the fuel lean-to-stoichiometric mixture due to the decrease in the flame thickness. Therefore, flame instability tends to increase at the lean-to-stoichiometric region. The increase in CO_2 in biogas by 10%, increases the Lewis number (L_e) value by 3.6% to 4.6%. The diffusive-thermal instability was dominant for all biogas-N_2O mixtures, as L_e was less than unity throughout the entire equivalence ratio range. The thermal reaction of N_2O decomposition is the most significant reaction in biogas-N_2O combustion at lean mixtures of ϕ = 0.6 and ϕ = 0.8. The LBVs of biogas-N_2O mixture revealed a considerable enhancement at the lean equivalence ratio of 0.8 compared to the other biogas-air mixtures with H_2 addition. The effect of nitrous oxide as an oxidizer on biogas detonation characteristics is studied numerically using Chemical Equilibrium Applications (CEA) and compared with other oxidizers. Mixtures with N_2O oxidant revealed 32% and 16% higher detonation pressure and detonation Mach number, respectively, at ϕ = 0.6, compared to that of mixtures with pure oxygen oxidant. Overall, employing N_2O oxidant has enhanced the fundamental combustion characteristics of biogas significantly, which may lead to the use of biogas as a clean fuel in commercial applications that demand high combustion rates, such as power generation and aerospace.

ABSTRAK

Biogas merupakan sumber tenaga alternatif yang berpotensi dengan kesan alam sekitar yang rendah. Walau bagaimanapun, aplikasi praktikal biogas agak terhad kerana kewujudan karbon dioksida (CO_2) yang bertindak sebagai bahan pencair yang mengurangkan nilai kalori dan kadar pembakaran biogas. Nitrous oksida (N_2O) dikenali sebagai agen pengoksidaan yang kuat untuk aplikasi pendorong yang boleh meningkatkan kadar pembakaran biogas. Walau bagaimanapun, terdapat kekurangan kajian yang menyelidit ciri asas pembakaran biogas-N_2O. Matlamat kajian ini adalah untuk mendapatkan gambaran tentang ciri-ciri pembakaran asas campuran biogas-N_2O dari segi halaju pembakaran laminar dan kestabilan nyalaan. Dalam kajian ini, nyalaan api yang mengembang secara sfera berikutan penyalaan secara berpusat dalam kebuk pembakaran isipadu malar (CVCC) digunakan untuk menyiasat halaju pembakaran laminar (LBV), ketidakstabilan hidrodinamik, dan ketidakstabilan terma resapan untuk campuran biogas-N_2O pada julat nisbah kesetaraan;ϕ, dari 0.6 sehingga 1.4 pada suhu 303K dan tekanan atmosfera. Dua mekanisme telah digunakan dalam perisian CHEMKIN-PRO 17 untuk menganggarkan ciri-ciri pembakaran yang diramalkan bagi campuran biogas-N_2O. Keputusan menunjukkan bahawa penurunan dalam LBV adalah lebih ketara dalam campuran tinggi bahan api berbanding dalam campuran rendah bahan api dengan kehadiran pencairan CO_2. Pengaruh lengkungan nyalaan pada bahagian hadapan nyalaan api didapati menjadi lemah pada campuran rendah bahan api ke campuran stoikiometri disebabkan oleh penurunan ketebalan nyalaan. Oleh itu, ketidakstabilan nyalaan api cenderung meningkat di kawasan campuran rendah bahan api dan stoikiometri. Peningkatan kandungan CO_2 dalam biogas sebanyak 10%, meningkatkan nilai nombor Lewis (Le) sebanyak 3.6% hingga 4.6%. Ketidakstabilan terma resapan adalah dominan untuk semua campuran biogas-N_2O, kerana Le adalah kurang daripada satu sepanjang julat nisbah kesetaraan keseluruhan. Tindak balas terma penguraian N_2O adalah tindak balas yang paling ketara dalam pembakaran biogas-N_2O pada campuran rendah bahan apiϕ = 0.6 dan ϕ = 0.8. LBV bagi campuran biogas-N_2O menunjukkan peningkatan yang ketara pada nisbah kesetaraan rendah bahan api 0.8 berbanding campuran biogas-udara lain dengan penambahan H_2. Kesan nitrus oksida sebagai pengoksida ke atas ciri letupan biogas dikaji secara berangka menggunakan Aplikasi Keseimbangan Kimia (CEA) dan dibandingkan dengan pengoksida jenis lain. Campuran dengan pengoksida N_2O menunjukkan tekanan letupan dan nombor Mach letupan masing-masing 32% dan 16% lebih tinggi, pada ϕ = 0.6, berbanding dengan campuran dengan pengoksidaan oksigen tulen. Secara keseluruhan, menggunakan pengoksida N_2O telah meningkatkan ciri-ciri pembakaran asas biogas dengan ketara, yang boleh membawa kepada penggunaan biogas sebagai bahan api bersih dalam aplikasi komersial yang memerlukan kadar pembakaran yang tinggi, seperti penjanaan kuasa dan aeroangkasa.

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B60	Biogas with 60% methane
B65	Biogas with 65% methane
B70	Biogas with 70% methane
B75	Biogas with 75% methane
B80	Biogas with 80% methane
BR	Blockage ratio
CEA	Chemical Equilibrium Applications
CH_4	Methane
CJ	Chapman Jouget
CO	Carbon Monoxide
CO_2	Carbon Dioxide
COD	Chemical oxygen demand
CPO	Crude palm oil
CVCC	Constant volume combustion chamber
DRM	Developed Reduced Mechanism
GHG	Greenhouse gasses
GRI	Gas Research Institute
ICE	Internal combustion engine
H_2	Hydrogen
LBV	Laminar Burning Velocity
LPG	Liquefied petroleum gas
N_2O	Nitrous Oxide
NOx	Nitrogen oxides
NUIG	National University of Ireland Galway
PAH	Polycyclic Aromatic Hydrocarbon
POME	Palm oil mill effluent
SCR	Selective catalytic reduction
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
a_{tt}	Tangential Strain Rate
β	Secondary Temperature Dependency
C	Capacitance
χ	Hydrogen addition 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
λ_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
Le_{eff}	Effective Lewis Number
L_{min}	Fuel Specific Stoichiometric Air Requirement
$(A/F)_{stoic}$	Stoichiometric Air-Fuel Ratio
M	Mach Number
Ma	Markstein Number
MW_{air}	Molecular Weight of Air
MW_{fuel}	Molecular Weight of Fuel
Pe	Peclet Number
Pe_c	Critical Peclet Number

ρ_b	Burned Gas Density
ρ_u	Unburned Gas Density
ϕ	Equivalence Ratio
R	Universal Gas Constant
R^2	Coefficient of Determination
Re_c	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_L	Laminar Burning Velocity
S_n	Stretched Flame Speed
S_L^0	Planer Speed of Flame Front
S_s	Unstretched Flame Speed

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Increasing global energy demand and emissions regulation has stimulated the research for clean and sustainable alternative fuels. While fossil fuels' combustion significantly increases greenhouse emission effects by emitting carbon dioxide, fossil fuel is still the dominant energy source for most industrial and agricultural activities. The increased power demand, depleting fossil fuel resources, and growing environmental pollution have led the world to explore seriously other alternative sources of energy. The basic concept of alternative energy relates to issues of sustainability, renewability, and pollution reduction (El Hawary *et al.*, 2016; Basha, Gopal and Jebaraj, 2009; Hosseini and Wahid, 2013).

Biogas is a clean source of energy that has great potential as a conventional fuel alternative. The fuel has a minimal environmental effect and has been used in many combustion applications. In contrast to the other natural resources including coal and fossil fuels, biogas is renewable and highly efficacious for reducing greenhouse gas emissions (Migiro, 2010). Biogas can be obtained by a process identified as the (anaerobic digestion) process. The anaerobic digestion method basically is a breakdown of organic matter with the absence of oxygen. Therefore, biogas originally comes from converting organic waste to a useful resource of energy. (Mihic, 2004; Xin *et al.*, 2013).

Biogas is a mixture of different gases consisting mainly of methane (CH_4) with some carbon dioxide (CO_2), carbon monoxide (CO) and traces of other gases. The contaminants and composition of biogas are varying depending on the composition of the feed stock as shown in Table.1.1. The proportion of CH_4 and CO_2 are the major features which affect the biogas' calorific value.

Table 1.1 Biogas composition and qualities (Chen *et al.*, 2015)

Component	Agricultural waste	Landfills	Industrial waste
Methane CH_4	50–80	50–80	50–70
Carbon dioxide CO_2	30–50	20–50	30–50
Hydrogen sulphide H_2S	0.70	0.10	0.80
Hydrogen H_2	0–2	0–5	0–2
Nitrogen N_2	0–1	0–3	0–1
Oxygen O_2	0–1	0–1	0–1
Carbon monoxide CO	0–1	0–1	0–1
Ammonia NH_3	Traces	Traces	Traces
Siloxanes	Traces	Traces	Traces
Water H_2O	Saturation	Saturation	Saturation

Nitrous oxide (N_2O) is often referred as the laughing gas owing to the euphoric influences during the breathing. It has been utilized in a range of combustion applications such as rocket engines, and internal combustion engines. N_2O generates higher O_2 content than air, allowing the engine to consume more fuel, providing higher intense combustion, which is desired particularly in automobile racing. Since N_2O provides several advantages over the other oxidizers, including being less toxic, stable at room temperature, convenient to stock, and generally safe to carry on aircraft, it may be utilized as a suitable oxidant in the rockets. Several recent investigations have been interested in studying N_2O since it is appropriate as monopropellant and bipropellant rocket system. Due to its unique properties, nitrous oxide can be used for a wide variety of space applications(Shen *et al.*, 2019).

When N_2O decomposition occurs, it unleashes both oxygen and nitrogen molecules. This breakdown results in an O_2 content of 36.36 %. On the other hand, just 21% of O_2 is obtained from air, with the remainder containing N_2 and some other inert gases, providing a12% lower O_2 content compared to N_2O. The additional content of oxygen aids combustion by reacting with hydrocarbon fuels to release CO_2 and H_2O, in addition to heat, which yields the combustion products to expand and generate higher pressure. Due to the exothermic decomposition of N_2O combustion into oxygen and nitrogen gas, it generates a greater temperature in the combustion engine which further enhances engine performance and efficiency, this is directly linked to the temperature variation in the unburned and the burned gases generated in the combustion chamber.

Combustion is the process of extracting energy from the chemical bonds of fuel molecules. Fuel molecules are oxidized by chemical interaction with oxygen during combustion, generating heat and creating specific combustion products. However, understanding the combustion properties of biogas is required to optimize biogas utilization while minimizing undesired combustion products (Moran and Shapiro, 2006)

The focus of this study is to investigate the laminar premixed biogas-nitrous oxide combustion. Such kind of combustion allows the fuel to be burned at stoichiometric and non-stoichiometric equivalence ratios. Under the lean condition, lower temperature combustion can be reached. Laminar burning velocity (LBV), Markstein length, flame stability, and flammability limit are among the properties frequently used to describe the combustion behavior of fuels. These fundamental characteristics are unique to each fuel since they are affected to some extent by fuel properties.

The rate at which the combustible mixture propagates into the reaction zone is known as laminar burning velocity. LBV has a significant effect on the development of pressure, particularly in a closed system. Further details and full description of laminar burning velocity and combustion characteristics will be discussed in chapter 2.

Markstein length is a characteristic that describes the impact of changes in flame structure owing to stretching to the speed of the flame. Markstein length is affected by the chemical and transport properties of the reacting mixtures. It can be used as an indicator of the impact of the stretch of the flame on the laminar burning velocity (Li *et al.*, 2014).

1.2 Problem Statement

Although there are considerable studies on utilizing biogas as an alternative source of fuel, the low calorific value of biogas owing to the existence of CO_2 has restricted the usage of biogas in some commercial industries that demand high combustion rates, such as aerospace and propulsion industries.

Carbon dioxide concentration may have an adverse influence on biogas combustion by reducing the calorific value, flame velocity, and flame stability. CO_2 could absorb a portion of the heat from combustion, that reducing the temperature of the combustion and the flame propagation (Hinton and Stone, 2014).

Laminar burning velocity determination is essential to evaluate the impact of CO_2 proportion on biogas combustion. (Fischer and Jiang, 2015) revealed that CO_2 could have a negative influence on biogas reactivity particularly at percentages more than 50% due to the significant drop in CO mass fraction, which would have an impact on certain important reactions in the combustion process. Therefore, a subsequent reduction in biogas LBV is expected with higher CO_2 percentage.

Nitrous oxide is a potent oxidant that is widely used in rocket propulsion systems. It can produce a substantial amount of energy due to the positive enthalpy of formation of N_2O decomposition reaction which can release 82 kJ/mol (Lin *et al.*, 2018). Thus, as revealed in equations 1.2 and 1.3, N_2O can be a further potent oxidizer than pure oxygen, where the combustion of one mol CH_4 with N_2O produces about 30% more energy than of 1 mol CH_4 with O_2 at stoichiometric conditions (Lin *et al.*, 2018).

$$N_2O \rightarrow 1/2O_2 + N_2,\ \Delta H\text{^}{}^\circ C = -82 kJ/mol \qquad (1.1)$$

$$CH_4 + 4N_2O \rightarrow CO_2 + 2H_2O + 4N_2,\ \Delta H\text{^}{}^\circ C = -1219 kJ/mol \qquad (1.2)$$

$$CH_4+2O_2 \rightarrow CO_2+2H_2O, \Delta H\text{^°}C = -890 kJ/mol \quad (1.3)$$

Therefore, N_2O is usually utilized to boost the engine performance of race vehicles as well as used as an oxidant for rocket propulsion (Lin *et al.*, 2018). Thus, N_2O can be used as a potential oxidizer to improve the biogas' combustion rate.

Yet, studies on the combustion characteristics of Biogas-N_2O are still scarce, which represents a research gap. Therefore, this study aimed at gaining insight into the fundamental combustion characteristics of biogas-N_2O mixtures in terms of laminar burning velocity and flame stability at various ranges of equivalence ratios, in addition to conducting a comparison between Biogas-N_2O and Biogas with hydrogen addition in terms of laminar burning velocity.

1.3 Objectives of the Study

The objectives of this study could be summarized as follows:

1. To characterize experimentally and numerically the flame propagation characteristics of biogas-nitrous oxide mixture (with varying CH_4/CO_2 content) at initial conditions of 1 atm. and 303K in terms of laminar burning velocity.
2. To estimate the impact of hydrodynamic, diffusive-thermal instability and CO_2 variation on biogas-N_2O combustion.
3. To establish a comparison of combustion characteristics of biogas-N_2O, biogas-air, as well as biogas-air with hydrogen addition in terms of laminar burning velocity.

4. To determine the ideal-detonation characteristics of biogas-N_2O, biogas-air, and biogas-oxygen in terms of detonation pressure, detonation velocity, detonation temperature, and Mach number.

1.4 Scope of the Study

The scope of the current study covers:

(a) Experimental study of the spherical premixed laminar burning velocity of biogas and nitrous oxide mixture with varying CO_2 percentage for two different mixtures of biogas (75%CH_4/25%CO_2)-N_2O and (65%CH4/35%CO_2)-N_2O using outwardly propagating spherical flame approach at different equivalence ratios (ϕ =0.6 – 1.4) using constant volume combustion chamber (CVCC).

(b) Numerical study by using ANSYS CHEMKIN-PRO ver.17 to determine the predicted laminar burning velocity of biogas-N_2O mixtures and to compare with the experimental findings.

(c) Investigation of the effect of hydrodynamic and diffusive-thermal instability of biogas-N_2O combustion at different equivalence ratios (ϕ =0.6 – 1.4) using constant volume combustion chamber (CVCC).

(d) Comparison between the experimental results of the premixed laminar burning velocity of biogas-N_2O and H_2 addition biogas with different percentages of H_2 addition.

1.5 Significance of the Study

The main purpose of this thesis is to investigate the flame propagation characteristics and the flame stability of biogas-N_2O as a potential mixture to enhance the combustion characteristics of biogas. This study can aid to increase the biogas' performance in some industries which demand high rate of combustion such as propulsion and detonation industries. According to the World Biogas Association, biogas can reduce global climate change emissions by 20%, and it can become a global sustainable industry worth $1.3 trillion. This study aimed to open up new prospects for widening the biogas utilization in aerospace and propulsion applications due to the various advantages of nitrous oxide as an oxidizer which can be stored as a liquid phase (~745kg/m^3) with a vapor pressure of ~52 bar at 20 °C and it can decompose at adiabatic decomposition temperature of about 1640°C (Zakirov, Richardson, *et al.*, 2001; Zakirov, Sweeting, *et al.*, 2001; World Biogas Association, 2021).

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LIST OF PUBLICATIONS

1. Effect Of Nitrous-Oxide on Laminar Burning Velocity, Hydrodynamic, And Diffusive-thermal Instability of Biogas Combustion. (2022). Journal of Thermal Analysis and Calorimetry. https://doi.org/10.1007/s10973-022-11408-2.

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