Full Length Article

Experimental and computational investigation of the laminar burning velocity of hydrogen-enriched biogas

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ABSTRACT

Experimental investigation of the adiabatic laminar burning velocity (LBV) of mixtures representative of biogas and hydrogen-enriched biogas was carried out using heat flux method (HFM). To prepare samples of biogas for testing, pure methane (CH_4) was diluted with 5–50 percent carbon dioxide (CO_2). Thereafter, some of the biogas samples were enriched with either 20% or 40% hydrogen (H_2). The combined effect of dilution with CO_2 and enrichment with H_2 on the LBV of CH_4 at different equivalence ratios were studied experimentally using HFM and computationally using ANSYS Chemkin-Pro® with GRI Mech. 3.0 and San Diego reaction mechanisms. The experimental results indicate that dilution with CO_2 reduces the LBV of richer mixtures more significantly than that of leaner or stoichiometric mixtures. However, when the biogas samples were enriched with H_2, a significant rise in the LBV was observed for fuels with higher content of CO_2. Sensitivity analyses of mass flow rate using GRI Mech. 3.0 revealed that with increased CO_2 concentration, the sensitivity coefficient of $H + O_2 \leftrightarrow O + OH$ (R38) increases significantly and that of $OH + CO \leftrightarrow H + CO_2$ (R99) decreases slightly. The sensitivity coefficients of $H + O_2 + H_2O \leftrightarrow HO_2 + H_2O$ (R35) and $H + CH_3(+M) \leftrightarrow CH_4(+M)$ (R52) are largest for fuel with the maximum percentage of CO_2. Further, with an increased H_2 concentration in biogas mixtures, the sensitivity coefficient of $H + O_2 \leftrightarrow O + OH$ (R38) increases, and that of $OH + CO \leftrightarrow H + CO_2$ (R99) decreases. Additionally, the sensitivity coefficients of reactions $H + O_2 + H_2O \leftrightarrow HO_2 + H_2O$ (R35) and $H + CH_3(+M) \leftrightarrow CH_4(+M)$ (R52) increases with increased H_2 concentration. The combined effect of CO_2 and H_2 on the rate of consumption of CO via R99 for all tested fuels having $CH_4:CO_2$ from 1:1 to 4:1 was predicted computationally. The rate of consumption of CO was least for $CH_4:CO_2 = 1:1$ and the peak value of the rate of consumption increases with decreasing CO_2 concentration irrespective of H_2 concentration in the fuel.

1. Introduction

According to the World Energy Council, the World's per capita primary energy demand will peak before 2030 [1]. At the same time, combustion devices will remain one of the most significant sources of environmental pollution. Majority of the conventional fuels (e.g., diesel, petrol, etc.) used currently have high global warming potential (GWP) due to their long carbon cycles and emit high exhaust emissions. The greatest challenge for researchers is to limit the global average temperature rise below 2 °C [2] and develop sustainable alternative sources of energy to reduce greenhouse gas emissions [3].

An important candidate for non-conventional cleaner fuel technology is hydrogen (H_2) among biogas, natural gas, producer gas and others. These gaseous fuels emit less pollutants and readily form homogenous mixtures in contrast to conventional liquid fuels [4]. Biogases that are produced by the biochemical decomposition of biomass (e.g., animal dung, agro-wastes, and municipal solid wastes), are primarily a mixture of 45–60% CH_4, 40–55% CO_2 along with some trace amounts of H_2S, N_2, CO, O_2, and water vapor [5]. Biogas has a low heating value due to its high CO_2 content. The maximum LBV of biogas (60% CH_4 - 30% CO_2 - 0.18% CO - 0.18% H_2) in the air at ambient temperature is about 25 cm/s, while that of liquefied petroleum gas (30% C_3H_8 - 70% C_4H_{10}), which has LBV of 38.25 cm/s and of H_2 is 275 cm/s [4]. At 1 bar, 15 °C the LHV of biogas, LPG, natural gas, and H_2 with above concentrations are 17 MJ/kg, 45.7 MJ/kg, 50 MJ/kg and 120 MJ/kg respectively [4]. Besides the fact that the biogas has a low calorific value, it still finds applications in cooking, refrigeration, electricity generation, transportation, and lighting especially in the Indian context due to the large cattle population [6]. Further, biogas may be used to substitute diesel in diesel engines, saving up to 70–80% diesel [6]. On the other hand, H_2 as a fuel has the following properties to its advantage: low density, high flammability, high mass-based

calorific value, high reactivity and zero emissions. H_2 not only improves extinction and flammability limits of lean hydrocarbon-air mixtures but also enhances the combustion efficiency of lean and low calorific value fuels. Due to these characteristics, H_2 has significant potential as a clean energy carrier for a broad range of applications including power generation and transportation.

For premixed combustion, important characteristics are LBV, adiabatic flame temperature (AFT), ignition delay, flammability limits and quenching distance. The primary objective of this paper is to study the effects of CO_2 dilution and H_2 enrichment on the LBV and flame structure of CH_4-air mixtures, specifically when it is diluted and enriched simultaneously.

The study by Kishore et al. [5] shows that the LBV of CH_4 is reduced more by CO_2 dilution than N_2 dilution. Additionally, the peak LBV for CH_4-air mixtures occur near the stoichiometric condition when diluted with 20%, 40% or 60% CO_2 by volume. Mohammed et al. [7], Xu et al. [8] and Gascoin et al. [9] reported that the replacement of N_2 with CO_2 decreases OH concentration, peak flame temperature and NO concentration in emissions due to the high specific heat of CO_2. Various studies [10–14,50] reported that the presence of CO_2 in any fuel tries to capture H radicals via reaction $CO + OH \leftrightarrow CO_2 + H$, which decreases the reaction rate, the flame temperature, and the LBV. Jahangirian and Engeda [15] concluded that for a fixed hydrocarbon content (i.e., CH_4 = 50%), replacing CO_2 with N_2 and H_2O led to an increase in H radical concentration and observable NO emissions. Park et al. [17] observed that the peak burning velocity shifted towards stoichiometry (equivalence ratio, $\phi = 1$) with an increased CO_2 concentration in the fuel mixture. These observations were similar to Chan et al. [16] and Yang et al. [18]. The probes of Lee et al. [19] and Hu et al. [20] reported that with increased CO_2 dilution in different gaseous fuels, the reduction in LBV and temperature was due to dilution, thermal, chemical and radiation effects. H_2 enrichment effects on CH_4 and biogas fuels were also analyzed in previous studies [21–28,31,49]. According to these, the heat release rate, flammability, flame stability, flame temperature, reaction energy, LBV, and active radicals concentration (H, O, and OH) increases due to H_2 enrichment. However, the CO emission decreases and peak LBV shifts towards the rich mixture side. Wei et al. [25] numerically studied the chemical kinetics of the premixed laminar biogas flames in the presence of H_2 using GRI Mech. 3.0 [43]. They observed that the CO_2 dilution reduces the global heat release rate due to the dilution, thermal and chemical effects. However, the addition of H_2 in biogas improves the global heat release rate significantly. Zhen et al. [26] studied the stability and thermal emission characteristics of the biogas-H_2 mixture using Bunsen burner method (for $400 \leq Re \leq 800$ and $0.8 \leq \phi \leq 1.2$) and observed that the LBV of biogas-air mixtures increases monotonically with H_2 addition. Further the peak LBV shifts from $\phi = 1$ to $\phi = 1.2$. Xie et al. [32–33] reported that the key free radicals like H and OH play an important role in chemical kinetics of the diluted fuels. Pizzuti et al. [34] conducted a detailed review on LBV and flammability limits of biogas and concluded that when biogas is enriched with fuels like H_2, natural gas and propane, the lean flammability limit and LBV increases. Cardona et al. [29] measured the LBV of biogas (66% CH_4 - 34% CO_2) and a biogas/C_3H_8/H_2 mixture (33% CH_4 - 17% CO_2 - 40% C_3H_8 - 10% H_2) with normal and oxygen-enriched air at 0.828 atm and 298 K for varying equivalence ratios. They observed that the LBV of CH_4 and biogas/C_3H_8/H_2 mixture differ by about 10%, thus making them interchangeable. Mameri and Tabet [30] found that by enriching biogas diffusion flames with H_2, the mixture becomes more reactive leading to increase in reactive radical concentrations, heating value and flame temperature.

Porpatham et al. [35] experimentally examined an SI engine fueled by H_2-enriched biogas (5%, 10%, and 15%), and reported that even small quantities of H_2 addition to biogas may enhance the thermal efficiency and the power output of the engine with an appreciable

reduction in the hydrocarbon emissions. Donohoe et al. [36] reported that an increase in temperature and pressure results in a decrease in ignition delay for H_2-enriched CH_4 and natural gas at atmospheric and high pressures.

Abundant literature is available on LBV measurements of CO_2 diluted CH_4 using different measurement techniques. However, not much data is available for mixtures when CH_4 is diluted and enriched simultaneously with CO_2 and H_2 respectively. The literature [12,28–31,35,49] suggests that the combustion characteristics of low calorific value fuels like biogas can be significantly improved by enriching them with some high-grade fuels like H_2, as the fuels with low energy content acquire some of the combustion properties of H_2 when suitably enriched. In the present work, an attempt has been made to improve the combustion characteristics of biogas fuels by enriching them with H_2. Also, the flame structure of CO_2-diluted and H_2-enriched CH_4-air mixtures are studied in detail. For experimental work, the heat flux method due to de Goey and co-workers [37–41], is used for measuring the adiabatic laminar burning velocity of multicomponent gaseous fuel mixtures at 1 bar and 298 K.

2. Experimental and computational details

For experimental investigations, a flat flame burner similar to de Goey and co-workers [37–41] and Kishore et al. [5] was used. The setup was developed by our group, details of which can be referred from [5,56–57], is used for experimental work. However, a brief description is presented here. The setup consists of a circular; honeycomb holed, brass burner plate of thickness 2 mm and diameter 30 mm. The number of holes in the burner plate is approximately 1519 with diameter 0.5 mm and pitch of 0.7 mm. Mass flow controllers (Mfg. Alicat Scientific Inc., U.S) are used to control and regulate the mass flow rate of individual gases. K-type (chromel-alumel) thermocouples with a bead size of 0.3 mm are attached at different radius-angle locations in the burner plate to record the temperature. The thermocouple wires are insulated with teflon to prevent any heat interaction with the surroundings. The gas flow through the mass flow controllers were set and monitored using a PC (using FLOWVISION® Software).

The unstretched adiabatic LBV is determined using interpolation technique proposed by de Goey and co-workers [37–41]. This is done by plotting the parabolic coefficient as a function of unburned mixture velocity and identifying the mixture velocity where parabolic coefficient becomes zero through interpolation. A photograph of the experimental facility is shown in Fig. 1. The inlet velocity of the fuel-air mixture, calculated through the volumetric flow rate of the gases passing through the burner orifice, is metered by the mass flow controllers (MFCs). The mass flow controllers used in the present work have an accuracy of ± 0.4% of the reading ± 0.2% of the full scale. The mass flow controllers are pre-calibrated to handle 29 different gases. The gas to be metered by an MFC is chosen using the gas selection menu, of Flow Vision® software supplied by the manufacturer. The MFCs can be connected to a RS232 communication port and controlled from the PC using the software. The MFCs meter the appropriate amount of each gas as per the set requirement and send it to the mixing tube. The mass flow controllers were calibrated before starting experiments. In order to minimize the uncertainty, all the mass flow controllers were used above 10% and below 90% of their respective range specified by the manufacturer.

For computational analysis, ANSYS Chemkin-Pro® [42] with GRI Mech. 3.0 [43], San Diego [46] and for some cases USC II [44] reaction mechanisms were used. The experiments and simulations were conducted at 1 bar, 298 K with equivalence ratio ranging from 0.7 to 1.4. The simulations accounted for multi-component transport. The Soret effect was used for the fuels containing H_2. The adaptive grid parameters (GRAD = 0.025 and CURV = 0.1) were suitably selected to obtain grid-independent values of LBV.