Study on the characteristics of hydrogen-enriched biogas co-flow flameless combustion

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Abstract: In this paper, the effects of biogas hydrogen enrichment on the characteristics of co-flow nonpremixed flameless combustion and pollutant formation are developed experimentally and numerically. Biogas (40% CO₂ and 60% CH₄) is applied in the flameless furnace as a fuel and the combustion stability, temperature uniformity inside the chamber and NO_x formation are studied when hydrogen is added to the components of biogas form 2% up to 10% by volume. A numerical approach by Flamelet combustion and turbulent standard k– ϵ model is applied. The results indicate that stability of the biogas flameless regime and uniformity of the temperature within the chamber could be ensured by adding just 2% hydrogen to the biogas components. In the further hydrogen concentrations, the peak temperature increases however compared to 2% hydrogen concentration, the mean temperature throughout the chamber reduces. The rate of NO_x formation mitigates when hydrogen is added to the biogas components up to 4% by volume. However, from 4% to 10% additional hydrogen, the rate of NO_x constitution intensifies due to high peak temperature.

Key-Words: Biogas, Hydrogen, Flameless combustion, Flamelet, NO_x

1 Introduction

Today, fossil crisis and pollutant formation cause serious problems, which challenge the conventional fuel utilization patterns. To address these dilemmas, a high quality fuel named hydrogen energy has been introduced to replace the traditional one. However, storage and transport of pure hydrogen is difficult due to its high flammability and diffusivity nature. Therefore, hydrogen enrichment of various gaseous fuels like methane (CH₄) and propane (C_3H_8) have been developed. Hydrogen containing fuels (HCFs) could be achieved by biomass gasification to generate electricity by direct combustion [1]. Although the stability of flame is ensured due to high flammability of hydrogen, NO_x may be largely formed via thermal-NO_x mechanisms due to the high adiabatic flame temperature of the HCFs [2]. By using premixed combustion systems, NO_x could be suppressed in very lean mixtures, however this could produce flashback phenomena and stability problems. Meanwhile, biogas generated from anaerobic digestion (AD) of biomass is a renewable source of energy for combined heat and power (CHP) generation. Likewise, environment could be preserved from greenhouse gases (GHGs) emissions by capturing biogas from waste materials [3]. However, industrial utilization of pure biogas is not

feasible due to the low calorific value (LCV) of biogas and biogas should be upgraded to remove impurities such as CO₂ and H₂S [4]. In recent decade, rapid industrialization along with fossil fuel depletion, GHGs generation and global warming (GW) have brought great attention to academic societies and industrial sections to reduce pollution and increase the combustion efficiency. Flameless combustion is a promising technology for replacing the conventional combustion technologies due to its capabilities to mitigate fuel consumption and control pollutants formation simultaneously. Flameless combustion technology could decrease the flame temperature and reduce NO_x concomitantly and offering uniform temperature field. While reducing the residence time and the oxygen concentration in the high temperature reaction zone, the temperature gradient and the hot spots in the furnace are suppressed [5]. Thus thermal-NO_x can be reduced to a low level. Flameless combustion is generally achieved when diluted oxidizer is preheated over the auto-ignition temperature of the fuel. Fuel oxidation process is distributed across a large volume of the combustion chamber rather than limited to a flame front, subsequently elimination of peak temperatures and thermal NO_x suppression is expectable in the flameless mode. Flameless combustion technology

is suitable for those industries that high and uniform temperature distribution inside the combustion furnace is vital (e.g., in the glass making process, steel and ceramic industries) [6]. Furthermore, since combustion of LCV fuel in the flameless mode is well sustained, biogas released from waste products could be applied for CHP generation purposes in the flameless chambers and waste-to-well strategy could be adopted properly by biogas flameless technology [7]. Indeed, flameless combustion of hydrogen-enriched biogases could be interesting due to specific properties of hydrogen such as very high heating value, laminar flame speed and adiabatic flame temperature and combustion could be sustained when hydrogen is added to the fuel [8]. In the other hand, depend on the biogas feedstock, hydrogen could be one of the ingredients of biogas and the percentage of hydrogen could be increased by adding some additives [9]. Hence, study about behavior of hydrogen-enriched the biogas combustion is more highlighted in terms of management of biogas resources. Recently, many experimental and numerical investigations have been performed to clarify various nuances of hydrogen-enriched fuel in the flameless combustion method. Derudi et al. [10] successfully utilized the byproduct gases of the coke making process which is called coke oven gas (COG) (a mixture of CH₄ and H₂ with 40% and 60% by vol respectively) in flameless furnace. The numerical kinetic results of different CH₄/H₂ mixture flameless combustion simulated by Yu et al [11] were in agreement with the experimental results. Sabia et al. [12] pointed out that addition of small amounts of H₂, reduces the thermo-kinetic instabilities of CH4 flameless combustion. Since modeling of turbulence and chemistry interactions plays crucial role to figure out various aspects of flameless combustion phenomena, numerical investigations have attracted more attentions. In traditional combustion, the fuel oxidation processes take place instantaneously and the reaction rate is governed by turbulent mixing. However, in flameless mode, the low reactants concentration leads the combustion system to an augmentation of the chemical timescales. The turbulent mixing timescales decreases due to enhance turbulence levels emerged by extremely high speed of reactants. Consequently, the chemical and turbulent timescales become similar and the Damkohler number (the ratio of turbulent to chemical timescale) approaches unity [13]. In these circumstances both chemistry and mixing issues should be taken into account in the numerical analyses [14]. Christo and Dally et al [15] stipulated that molecular diffusion plays crucial role in the

simulation of a jet of CH₄/H₂ in hot co-flow (JHC) flameless mode. They compared different models of chemical kinetic, turbulence and combustion to anticipate their experimental data and concluded that the numerical results of Eddy Dissipation Concept (EDC) model are accurate for high percentages of oxygen in hot co-flow. They also claimed that the accuracy of EDC modeling with detailed chemical kinetics is more than global mechanisms. Parente et al. [16] claimed that the effects of molecular diffusion in the temperature of the flameless semi-industrial burner are negligible when internal exhaust gas recirculation is applied. The effects of various combustion models (EDC and Eddy Dissipation/Finite Rate (ED/FR)) and different kinetic mechanisms (GRI-3, DRM-19 and global) on CH₄ and CH₄/H₂ mixture combustions were investigated. Based on their numerical and experimental results, the role of a detailed chemistry approach in the modeling of flameless combustion structure is more highlighted. It was concluded that the role of molecular diffusion is depend on the way in which flameless mode is achieved. In an experiment done by Dally et al. [17], a jet of CH_4/H_2 was conducted to a hot co-flow oxidant stream in flameless conditions and the effects of various O_2 mass fractions of co-flow on the characteristics of combustion were investigated at different distances from the burner. The effects of various turbulence models (k $-\epsilon$ and the Revnolds stress model (RSM)) on the flameless combustion structure under EDC chemistry interaction were investigated bv Frassoldati et al. [18] and some differences in NO prediction between experimental and numerical results were reported. These discrepancies were attributed to the overestimation of the temperature in simulations. Ihme and See et al. [19] modeled MILD combustion by Large eddy simulations (LES) where presumed probability density function (PDF) was applied to simulate the turbulent chemistry interaction. It was found that equality of numerical results and experimental records significantly depends on the selection of appropriate boundary conditions. Moreover, LES method has great capability to predict the behavior of flameless combustion regime. The impacts of the fuel Reynolds number on the structure of the MILD flameless combustion was studied by Medwell et al. [20] by analyzing the distribution of temperature. CH₂O and OH in the furnace. Their results indicate that the augmentation of the fuel Reynolds number suppresses OH distribution and eliminate visible local flames. Moreover, higher mass fraction of CH₂O in low oxygen concentration condition is recorded due to reduction in reaction rates.

Oldenhof et al. [21] applied Planar Laser Induced Fluorescence (OH-PLIF), Laser Doppler Anemometry (LDA) and Coherent Anti-Stokes Scattering (CARS) to investigate the interaction of chemistry and turbulence in the several hot co-flow jet flames. They concluded that chemical reactions cause a minor effect on the velocity in these flames. Indeed, it was shown that in high jet Reynolds numbers the jet lift-off increases [22]. In combustion modeling of hydrogen-enriched fuel, it was claimed that the Arrhenius reaction rate increases due to the growth of the temperature, and the rate of O₂ consumption in the lean mixture is enhanced. It was found that in the combustion simulation of hydrogen-enriched gaseous fuel, fast chemistry models are superior due to their low computational cost. Indeed, the conserved scalar model as a subcategory of some other models such as flame sheet, laminar Flamelet and conserved scalar models with equilibrium chemistry is defined based on the relationship of the flame thermochemical characteristics as a function of the mixture fraction. However, some researchers claimed that due to the nature of chemical mechanisms of hydrogen-enriched fuel flameless combustion, ED/FR and EDC combustion mechanism should be taken into account and fast chemistry approaches like Flamelet and Eddy Dissipation Method (EDM) are not appropriate [23].

On the other hand, various chemical reaction mechanisms have been applied in hydrogenenriched gases fuel simulation. Ilbas et al. [24] employed only seven species and Yilmaz et al. [25] used GRI mechanism with 18 species. GRI2.11 was applied by Ravikanti et al. [26] and it was found the simulated results are in good agreement with the results of a reduced DRM-22 mechanism [27]. In the combustion simulation done by Frassoldati et al. [18], 600 reactions were implemented when 48 species were considered. Totally, a great variety of turbulence modeling, chemical reaction and interaction of these two have been introduced, however there is no universal suitable model for all turbulent combustion applications achieved. Each model shows several advantages and disadvantages and poses better performance only in specific applications. Reynolds averaged Navier-Stokes modeling (RANS) [28], large eddy simulation (LES) and direct numerical simulation (DNS) are the main methods have been applied for numerical investigation of non-premixed combustions. In turbulence modeling, despite the Reynolds Stress Model (RSM) yielding proper results for prediction of high strain rate flows and streamline curvature, its function is under question due to different results

issued by various researchers. In this paper the impacts of hydrogen addition on the stability of biogas flameless combustion and pollutant formation is investigated. Pure biogas (40% CO₂ and 60% CH₄) is applied as a fuel in a flameless chamber experimentally. The numerical model of biogas flow field and related chemical reactions are investigated. After model validation, hydrogen is added to the biogas and the impacts of such change in fuel composition on the temperature distribution in the furnace, flameless stability and pollutant formation is investigated in six cases: Case1 (H0): 0% H₂, 40% CO₂, 60% CH₄ Case2 (H2): 2%H₂, 40%CO₂, 58% CH₄ Case3 (H4): 4% H2, 40% CO2, 56% CH4 Case4 (H6): 6% H₂, 40% CO₂, 54% CH₄ Case5 (H8): 8% H₂, 40% CO₂, 52% CH₄ Case6 (H10): 10% H₂, 40% CO₂, 50% CH₄).

2 Experimental Setup

Fig1 demonstrates the experimental setup of the labscale hydrogen-enriched biogas flameless system.



Fig. 1 Experimental setup

The burner consists of a biogas/hydrogen inlet jet, mounted in the center which is surrounded by the holes prepared for high temperature diluted oxidizer inlet. The inlet diameter of the fuel nozzle and all oxidizer jets are 5 mm and the diameter of the exhaust is 50 mm. The diameter and the length of the cylindrical furnace are 264 mm and 600 mm respectively made of carbon steel. The inside diameter of the chamber is 150 mm after installation of refractory. Five holes are located at the top of the furnace in a specific distance from burner to record temperature and pollutants by K-type thermocouples and gas analyzer respectively. The inlet pressure and temperature of the biogas-hydrogen mixture and hot diluted co-flow oxidizer are 1.013×10^5 Pa, 300K and 900K respectively. The velocity of preheated oxidizer jet is kept constant at 90 m/s in the flameless mode in all cases. In this experiment the concentration of oxygen and nitrogen in the diluted oxidizer is 7% and 93% respectively.

In order to achieve flameless mode, three steps should be implemented. At the first step, furnace should be heated up and the temperature inside the chamber should be increased over the auto-ignition of the fuel. Therefore, combustion system is run in conventional mode. Since high temperatures could not maintained by conventional combustion of biogas, pure methane is applied to enhance the furnace temperature [29]. Second step which is called transient, is occurred when the temperature inside the whole of the chamber becomes significantly higher than biogas self-ignition (the mean temperature of the chamber is 1500K). In this circumstance, fuel is switched from methane to biogas. In the third step, which is flameless mode, fuel (biogas or hydrogen-enriched biogas) and preheated and diluted oxidizer (900K, 7% oxygen concentration) are injected to the furnace. In flameless step, the density of the oxidizer decreases dramatically due to preheating process (the density of air is 1.177g/l and the density of preheated and diluted air is 0.383 g/l), thus in order to maintain stoichiometric condition, oxidizer is injected to the furnace at very high velocity. Therefore, desirable turbulence circumstance for flameless mode is provided.

3 Numerical Study

A three-dimensional (3D) computational domain is simulated based on the experimental flameless chamber geometry. 3D simulation is done by ANSYS 14 using ANSYS Modeler to design the chamber and ANSYS Meshing to mesh the combustor [30]. To improve the convergence rate and scalar properties, mesh refinement is done, thus grid resolution for smooth flow representation is ensured. Due to symmetry, only one eighth of the furnace is simulated. The grid consists of 7769 nodes and 33798 elements. It is assumed that all the properties of a cell have an average value at the center of the cell. The second order upwind scheme is selected to calculate the temperature, velocity and pressure through the governing equations with coupled algorithm. A mass-weighted averaging method is set to interpolate the velocity values of cell-center to face values. The temperature of fuel and air inlet and pressure outlet is 300 K, 900 K and 1.013×10^5 Pa respectively and free stream turbulence is set at 5%. The residual energy

equation should drop below 10⁻⁶ and for all other variables it is set at 10^{-3} to ensure the convergence of the solution. The swirl velocity of components is neglected in the steady-state computational fluid dynamic (CFD) simulation and stoichiometric equivalence ratios (Φ =1) is considered. The density of biogas includes 40% CO₂ and 60% CH₄ in the standard temperature and pressure is considered 1.2146 g/l. The records of numerical solution with 7769 nodes and 33798 elements are in good agreement with the experimental results. The grid independent of the simulation is tested by changing the number of meshes to the finer meshes. In this simulation the number of elements was adopted 33798, 68453 and 100356 respectively. These adoptions were motivated by this fact that the most significant conformity to the experimental measurements could be achieved when the mesh is so fine. However meaningful changes were not observed in the records and the grid independent of solution is confirmed.

3.2 Turbulence Modelling

The standard $k-\varepsilon$ model is applied to model hydrogen-enriched biogas flameless combustion with the laminar Flamelet combustion model. In the standard $k-\varepsilon$ model, k and ε are modeled with following transport equations [29]:

$$\frac{\partial}{\partial t}(\rho k) + \frac{\partial}{\partial x_i}(\rho k u_i) = \frac{\partial}{\partial x_j} \left[\left(\mu + \frac{\mu_t}{\sigma_k} \right) \frac{\partial k}{\partial x_j} \right] + P_k + P_b - \rho \varepsilon - Y_M + S_k \tag{1}$$

$$\frac{\partial}{\partial t}(\rho\varepsilon) + \frac{\partial}{\partial x_i}(\rho\varepsilon u_i) = \frac{\partial}{\partial x_j} \left[\left(\mu + \frac{\mu_t}{\sigma_{\varepsilon}} \right) \frac{\partial k}{\partial x_j} \right] + C_{1\varepsilon} \frac{\varepsilon}{k} (P_k + C_{3\varepsilon} P_b) - C_{2\varepsilon\rho} \frac{\varepsilon^2}{k} + S_{\varepsilon}$$
(2)

where μ_t , P_k and Y_M represent turbulent viscosity, turbulence kinetic energy production and the contribution of the fluctuating dilatation in compressible turbulence to the overall dissipation rate respectively which are described by:

$$\mu_{t} = \rho C_{\mu} \frac{k^{2}}{\epsilon}$$
(3)

$$P_{k} = -\rho \overline{u'_{l} u'_{j}} \frac{\partial u_{j}}{\partial x_{i}}$$
(4)

$$Y_{M} = 2\rho \varepsilon M_{t}^{2}$$
(5)

3.2 Combustion Modelling

In the literature review it was pointed out that in the most of the previous publications, simulation of flameless combustion with EDC showed better results. However, Damköhler number distribution (D_a) , which represents the flow to chemical time-scale ratio, is effective for combustion modeling selection. Large Damköhler value corresponds to mixing controlled flames and low Damköhler value

indicates slow chemical reactions. It means that reactants and products are quickly combined by turbulence and the combustion system behaves like a perfect stirred reactor. Therefore, Damköhler number should be taken into consideration to set proper flow and chemical time-scales. Damköhler (D_a) number represents the behavior between mixing and reaction in a system, given by the ratio of a mixing or flow time-scale to a chemical timescale (τ_f/τ_c) [31]. Application of fast chemistry approaches in the numerical set up are appropriate when $D_a > 1$ [32]. In the present flameless furnace the mixing time is long and chemical reaction time is short (due to biogas utilization) and D_a is greater than one because of the specific geometry of the furnace (the distance between the centerline of fuel and oxidizer inlet is 52.5 mm). Therefore, application of fast chemistry approaches indicates better results in this geometry [33]. Using detailed chemical mechanisms, FLUENT can calculate laminar opposed-flow diffusion Flamelet for nonpremixed combustion. The laminar Flamelet are then set in a turbulent flame using statistical PDF methods. The sensible chemical kinetic impacts can be incorporated into turbulent flames by using the laminar Flamelet approach. The chemistry can be preprocessed and tabulated, offering enormous computational savings. For developing the Flamelet library, GRI3.0 mechanism includes 325 reactions and 53 species is employed.

3.3 Radiation Modelling

In the simulation of turbulent combustion, prediction of radiative heat transfer is very important. If an accurate radiative heat transfer method is not employed, notable discrepancies could be found between experimental results and numerical records in terms of pollutant formation and combustion characteristics. Prediction of NO_x formation is sensitive to the trend of chamber temperature, thus over prediction of NO_x formation occurs if the radiative heat loss is not taken into consideration. Discrete ordinates (DO) radiation model is used in this simulation because of its reasonable computational cost. DO is widely used in such similar computational investigation with no significant error [34].

3.4 NO_x formation

 NO_x reduction in combustion process of industrial sectors is necessary to control GW, acid rain, smog, ozone depletion and GHGs effects. NO_x is usually formed due to the reaction of nitrogen and oxygen within a locally high temperature region. The

environment and public health can be risked by increasing NO_x. At extremely high temperatures, N₂ and O₂ can react through chemical mechanisms called Zeldovich formulation. Thermal NO_x formation is quickened at high temperatures and the rate of formation increases quickly with increasing temperature. Prompt or Fenimore NO_x formation occurs in fuel rich conditions and it was found that the prompt NO_x formation increases near equivalence ratio of 1.4 [35]. N₂O-intermediate mechanism occurs in fuel lean, elevated temperature and low pressures. It was claimed that NO_x formation by N₂O mechanism is responsible for the main NO_x constitution in flameless mode. The calculation of thermal NO_x, prompt NO_x formation and N₂O-intermediate is done from global model presented in Ref [36].

4 Results and Discussion

When hydrogen is added to the biogas composition the density of the fuel decreases, therefore the mass flow rate of the fuel should increase to maintain stoichiometric condition. In the presence of 2% hydrogen in biogas components, the structure of biogas flameless mode changes and the maximum temperature increases. Indeed, the distribution of temperature inside the flameless chamber becomes more homogeneous. In the other word, the peak temperature shrinks and moves further away from the burner. In the further hydrogen concentrations, the peak temperature increases however compared 2% hydrogen concentration, to the mean temperature reduces throughout the chamber. Moreover, the temperature distribution within the flameless chamber is more uniform when just 2% hydrogen is added to biogas components. Fig2 depicts the contour of temperature inside the chamber in the four cases and experimental photos. Also, Fig3 display the impacts of hydrogen addition on the axial temperature distribution along with the flameless furnace.

Additional hydrogen changes the density of biogas and the flow rate of fuel mixture increases. The augmentation of temperature is not only attributed to the higher specific energy content of hydrogenenriched biogas, but to the decreased radiation losses caused by the reduction of dioxide carbon formation [37].



Fig.2 The contours of temperature distribution in the chamber



Fig.3 Effect of biogas hydrogen enrichment on axial temperature distribution along with flameless combustion furnace (numerical results)

The peak temperature of biogas flameless combustion increases when the hydrogen content of biogas is 2%. Further experiments with higher rate of hydrogen depict that the thickness of the peak temperature reduces. The peak temperature thickness reduction can be attributed to the augmentation of the mixing. The uniformity of temperature (T_u) defined based on Eq (6) is a value that illustrate whether temperature inside the chamber is homogenous or not [38]. Fig4 shows the value of T_u inside the chamber based on the numerical and experimental results.

$$T_u = 1 - \sqrt{\frac{1}{N} \sum_{i=1}^{N} (\frac{(T_i - \bar{T})}{\bar{T}})^2}$$
(6)

where T_u is temperature uniformity inside the chamber and N shows the total number of positions that temperature was measured, T_i is the temperature in the ith position and T is the mean of all the measured temperatures. The value of the T_u is between 0 and 1, where in perfect flameless combustion regimes the value of T_u approaches to one which demonstrates a perfectly uniform furnace. The trend of temperature uniformity in numerical results and experimental records is the same in all studied cases and the temperature uniformity in 2% hydrogen enrichment of biogas is more than other cases.



Fig.4 Temperature uniformity inside the chamber for experimental and numerical results

In the non-premixed combustion simulation, mixture fraction indicates the mixing rate. While the mixture fraction increases, it can be concluded that mixing does not take place properly. Fig5 shows the mixture fraction in axial and radial directions. When the hydrogen content of biogas is intensified, the radial spreading rate of mixture fraction is decreased which can be a sign of mixing growth.

The contour of NO_x constitution and the impacts of hydrogen enrichment on NO_x formation of biogas flameless combustion are presented in Fig6 and Fig7 respectively. From these figures it can be seen that the rate of NO_x formation decreases when hydrogen (up to 4%) is added to the biogas flameless regime. However, when the rate of hydrogen in the biogas content increases (from 4% to 10%) the rate of NO_x constitution intensifies. This phenomenon could be attributed to this fact that not only high temperatures but also oxygen concentration could play grate role on NO_x formation rate. Fig8 display oxygen concentration in the flameless chamber in various conditions. The rate of oxygen concentration in H2 and H4 is less than flameless combustion of pure biogas. Therefore low amounts of NOx formation in low level of hydrogen enrichment of biogas could be expected.



(B)

Fig.5 Effect of hydrogen enrichment on the (A) axial profile of mixture fraction and (B) radial profile of mixture fraction in biogas combustion

Fig.8 Oxygen concentration in hydrogen-enriched biogas flameless combustion (numerical results)

0.04

0.06

Axial distance [m]

H =% 0 H = % 2

➡ H = % 4
➡ H = % 6
➡ H = % 8
➡ H = % 10

0.10

0.08

0.008

0.006

0.004

0.002

0.02

O₂ mass fraction

5 Conclusions

LCV of biogas could be one of the most important barriers of biogas development which has been modified by hydrogen-enrichment strategy and using flameless combustion method. A numerical and experimental investigation of hydrogenenriched biogas flameless combustion has been presented. The effects of hydrogen enrichment on biogas co-flow flameless combustion were investigated flameless and characteristics. combustion stability and NO_x formation of pure biogas (40% CO2 and 60% CH4) and hydrogenenriched biogas (2-10% H₂ by volume) were studied. It was found that stability of the biogas flameless regime and uniformity of the temperature within the furnace could be ensured by adding just 2% hydrogen to the biogas components. In the concentrations, further hydrogen the peak temperature raises however compared to 2% hydrogen concentration, the mean temperature throughout the chamber decreases. The rate of NO_x formation mitigates when hydrogen is added to the biogas components up to 4% by volume. However, from 4% to 10% additional hydrogen, the rate of NO_x constitution increases due to high peak temperature. Numerical observations indicated the major role played by kinetic mechanisms and the turbulence chemistry interactions models in the simulation of the hydrogen-enriched biogas flameless combustion regime. It was found that the geometry of furnace and Damköhler number distribution (D_a), which represents the flow to chemical time-scale ratio, have grate impress on combustion model selection. Since in this project D_a is greater than one, Flamelet fast chemistry approach is superior. In particular, Flamelet with fast detailed chemistry provided results illustrating more homogeneous distribution of temperature as well as the reaction zone extension to a large portion of the available volume in the furnace, as it would be expected in hydrogen-enriched biogas flameless combustion.

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