Experimental and Computational Determination of LBV of Hydrogen Enriched Methane Mixtures

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ABSTRACT

One of the major causes of environmental pollution and ozone layer depletion is the emissions coming out of the combustion devices including industrial burners, automobile vehicles and household appliances. Most of the conventional fuels used now days have high GWP and ODP. So the greatest challenges among the combustion researchers and scientists are to develop some sustainable and non conventional sources of energy that possesses capability to replace the conventional ones. One of the important gaseous fuels in non conventional category is hydrogen, which is a cleaner fuel and reduces pollution enormously. In the present work, experimental & computational analysis of laminar burning velocity (LBV) of premixed gaseous fuels (primary focus on Hydrogen enrichment) was carried out. For experimental investigation the experimental set up available in Fuel and pollution lab of Indian Institute of Technology Delhi is used. Experiments were carried out on mixtures of methane- Air and Methane-Hydrogen-Air for wide range of equivalence ratios and compared with the computational results of PREMIX with full GRI-Mech 3.0 mechanism. Most of the experiments available in literature were carried out at 298 K. In the present work it has been tried to relate the effect of low temperatures on laminar burning velocity of mixtures. The experiments have been conducted at 1 bar pressure and around 292 Kelvin with equivalence ratio ranging from 0.8 to 1.2. Methane gas is enriched with hydrogen in varying proportions and the effect of hydrogen enrichment on its laminar burning velocity studied. The objective of the addition of hydrogen to methane was to increase its laminar burning velocity as well as to extend its lean flammability limits at lower ambient temperatures.

Keywords: *Laminar burning velocity, Heat flux, flat flame.*

1. INTRODUCTION

With the increasing population of the living world at an alarming rate, major concern is to meet the energy demand as well as protection of environment. According to the predictions of IEA [1], world energy demand will increase by more than 55 % by the year 2030. In this context researches are going on all over the world to provide clean, sustainable and renewable source of energy so that environment may remain protected.

In current scenario conventional fossil fuels such as coal, petrol and diesel are the key sources. These fuels are non-renewable, have long carbon cycle and high emission characteristics. The conventional fuels are diminishing day by day and are harmful for humans and environment. According to EIA IEO [1], 2007, world consumption of petroleum and other liquid fuels was around 83 million barrels oil per day (mbpd) in 2004 and will grow upto 118 millions in 2030. IEA Executive Director Maria van der Hoeven said in London at the launch of a World Energy Outlook Special Report, Redrawing the Energy-Climate Map, which highlights the need for intensive action before 2020 that "*This report shows that the path we are currently on is more likely toresult in a temperature increase of between 3.6 °C and 5.3*

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°C but also finds that much more can be done to tackle energy sector emissions without jeopardizing economic growth, an important concern for many governments"[2].

To overcome such problems, scientists have to move towards the alternative and sustainable fuels such as natural gas (primarily CH_4), biogas (CH_4-CO_2), and hydrogen etc. The potential sources of methane are landfills, industrial, agricultural, organic, animal and poultry wastes. A large amount of methane is produced during distillation of crude oil. Landfills are enormously available around us, which may be used for the production of methane as well as biogas. These fuels may be effectively used in automobiles, gas turbines, household kitchens, industrials burners, heating, cooling and other gaseous fuel related devices.

Hydrogen is one of the important alternative sources of fuel as it possesses almost all the qualities which are required by a good combustible fuel. H_2 is the most abundant gas found in this world and by principles it can be derived from various resources. If hydrogen is processed from a renewable resource, a carbon free society can be created. H_2 is an odorless, tasteless, non-metallic, non-toxic and highly combustible gas with a very low melting & boiling point. Molecular and thermal diffusivity of H_2 is very high. H_2 is pollutant free (Non GHG) and has heating value enrichment capability when mixed with a low grade fuel in suitable proportion. The laminar burning velocity of hydrogen is around 100-300 cm/s. Though H_2 is clean and very efficient but major problem in using H_2 as an alternative fuel is its production and storage. Some techniques like steam reforming of natural gas and electrolysis are used for H_2 production.

Laminar burning velocity, flame stretch, ignition delay, quenching distance, unburned gas mixture velocity and flammability limits are some key combustion characteristics of any premixed fuel/oxidizer mixture. If the flow of the fuel-oxidizer mixture is laminar, the flame speed of the premixed flames is dominated by the chemistry of the mixture. Designing combustion appliances for a variety of fuels requires basic idea of the associated effects with variation of different constituents of the fuel on its combustion characteristics. The LBV is a property of the mixture. Various researches reveal that for stoichiometric CH_4-Air mixture at 1 bar pressure and 298 K, the LBV comes around 36±1 cm/s [3]. In the present work, the laminar burning velocity of methane blended with hydrogen is determined at 292 K and compared with computational approach.

Abbreviations and definitions

LBV	laminar burning velocity
Mbpd	million barrels of oil per day
GHG	Green house gases
S_L	laminar burning velocity
H_f	volume fraction of hydrogen
Q	Net Amount of heat
Q_G	Amount of heat gain
Q_L	Amount of heat loss
MFC's	Mass flow controllers
GWP	Global Warming Potential
ODP	Ozone depletion potential

2. LITERATURE REVIEW

A number of research publications are available in which the LBV of methane, hydrogen and methane-hydrogen mixtures (Hythane) have been determined experimentally and computationally. But the LBV at lower temperatures (around 292 K) is not reported in literature. The effects of hydrogen addition on various fuels have also been studied. Zhang [4] reported that the laminar burning velocity, S_L, increases significantly with hydrogen fraction for all the equivalent ratios. Hydrogen enrichment increases the energy of reaction by which flame propagation speed and intensity of flame increases. Hu et .al. [5] carried out experiments at atmospheric pressure and room temperature and reported that the unstretched laminar

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burning velocity is increased and peak value of the unstretched laminar burning velocity shifts to a richer mixture side with the increase of hydrogen fraction. They observed that the enhancement of chemical reaction with hydrogen addition is due to increase of H, O and OH mole fraction in the flame. They also reported that a strong correlation is exists between burning velocity and maximum radical concentration of H and OH radicals in reaction zone of premixed flames as the reaction of H, O or H and OH evolves larger amount of energy nearly (about 418 KJ/kg). Increment in concentration of radicals results increase in order and speed of occurrence of reaction. Bougrine [6] studied the effect of H_2 substitution to natural gas and reported that addition of hydrogen enhances tendency of oxidation in burning of fuel which increases the flame burning speed. Chandra et al. [14] have conducted experiments on a stationary 5.9 KW CI engine converted into SI engine, with biogas as one of the alternative fuels for the converted engine (in place of CNG). They analyzed the engine performance parameters like brake power, specific gas consumption, thermal efficiency etc. on methane enriched biogas and reported that the biogas containing 95% CH_4 showed the engine performance similar to compressed natural gas (CNG). Their work justified the use of biogas as a strong alternative fuel for gasoline and CNG powered vehicles. Hence it can be concluded form the literature survey that addition of hydrogen increases the LBV and lean flammability limits of alternative sources of energy having low heating values such as biogas, producer gas etc.

3. HEAT FLUX EXPERIMENTAL SET UP USED IN THE PRESENT WORK

The heat flux method (HFM) originally developed by De Goey and co workers [7] is used in the present work. This set up is developed at Indian Institute of Technology, Delhi by Ratnakishore [11], refurbished and modified by Yadav [12]. The HFM technique demonstrates to measure the net heat loss from the flame to the burner plate and setting the unburned mixture velocity in such a way that no net heat loss to the burner occurs. Even this is an assumption as practically adiabatic condition is difficult to achieve.

The principle of heat flux method explains that to attain adiabatic condition (Where net heat transfer Q = 0). The burner plate's temperature is maintained around 85to 90 °C using a thermostatic hot bath which is covered with insulation from its sides to prevent any heat loss. The hot bath reduces the loss of heat from flame to the burner plate . A cooling water jacket circulation is also done to maintain the constant temperature of the incoming un burned gas mixture as per mixture requirement.

A cross section of 3 holes of the brass burner plate is shown in Figure 1, where the stream line of the gas flow are sketched on the left side showing that the flow is flattened by the pressure drop of flame front [11]. The total heat gain of the flame is defined as Q_G while the total heat loss is defined as Q_L. The net difference between the heat loss and the heat gain ($QL-Q_G$) is responsible for the temperature distribution that is observed in the burner plates by means of attaching thermocouples.

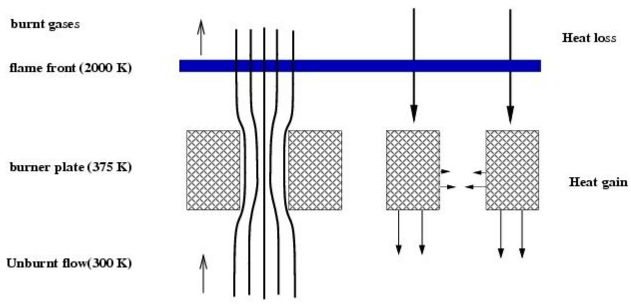

Fig.1. Principle of heat flux method [8]

The experimental setup used by us consists of a circular, honeycomb holed, brass burner plate, having 2 mm thickness and 30 mm diameter. The number of holes in the burner plate is approximately 1519 (after fabrication) with 0.5 mm diameter and 0.7 mm pitch. The mass flow controller (Mfg: Alicat Scientific Inc. U.S) are used to control and regulate the mass flow rate of associated gases. Five thermocouples (K-type Chromel-Alumel) with a bead size of 0.3 mm are attached to different radius-angle locations in the burner plate to read its temperature. The thermocouples are insulated with Teflon to prevent any interaction with surrounding. The input to the MFC's can be given through a PC (using FLOWVISION Software) and also the output can be displayed and recorded. The unstretched adiabatic laminar burning velocity is determined by interpolation technique proposed by De Goey and co workers [7]. This is done by plotting parabolic coefficient versus unburned mixture velocity and interpolating the condition where parabolic coefficient becomes zero.

4. NUMERICAL MODELLING

The Experimental Data was compared computationally using PREMIX Code [9] with full GRI Mech 3.0 mechanism [10]. The Laminar Burning Velocity at different equivalence ratios was computed for the various fuel configurations as shown in Table I

Table-1 fuel compositions for experimental study

Fuel Name	Fuel Composition used for experiments		
	CH_4 (%)	H_2 (%)	*OXIDIZER*
HY1	10	90	AIR
HY2	20	80	AIR
HY3	30	70	AIR

As the experiments were performed at 1 bar pressure, 292 K temperature and equivalence ratio ranging from 0.8 to 1.2, similar conditions were achieved computationally. The simulations were done on multi-component transport coefficient formulations. Soret effect (a phenomenon observed in mixtures of mobile particles where the different particles types exhibit different responses to the temperature gradient) was used. The following parameters were set for simulations: Maximum number of grids points allowed=1000, adaptive grid control based on solution gradient=0.02 and adaptive grid control based on solution curvature=0.1, the starting axial position= -2 cm and ending axial position=10 cm. The initial grid was based on temperature profile estimate. By selecting the gradient and curvature as above the LBV computations become independent of the grid [11].

V. RESULTS AND DISCUSSIONS

As a part of the present work, some experiments have been conducted on CH_4-Air and CH_4-H_2-Air at 292K and 1 bar pressure for different equivalence ratios. Before starting the experiments, the set up is validated by conducting some standard experiments on CH_4-Air mixtures at room temperature and atmospheric pressure. The obtained results showed nice trends with the available literature. Then the CH_4-Air experiments were conducted at 292 K and atmospheric pressure. For checking the repeatability of the experimental data some of the experiments were conducted four times on different days. The obtained results of various experiments were found to be in better agreement within their experimental uncertainty limits. While conducting the experiments extra precautions were taken to ensure the steady state attainment of the system. The hot bath was started 3 hours before start of the experiments and as the Digital Temperature Indicator shows a stable reading around 90°C, the experiments, and then only the experimental procedure started. Figure 2, 3 and 4 have been clicked using a high definition digital camera (Make SONY 16.1 Megapixels) while conducting the experiments.

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Fig. 2. Direct photograph of stoichiometric Hythane (10% H2-90% CH4)-Air mixture at p=1 bar and T=292 K

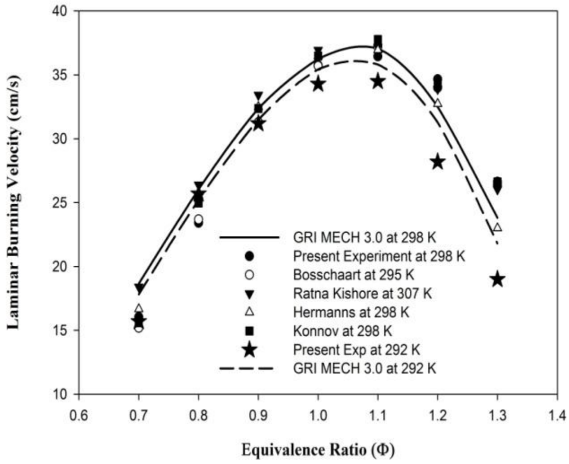

Fig.5. CH4-Air Validation experiments for stoichiometric Methane-Air mixture at 1 bar and 292 K compared with the results available in the literature.

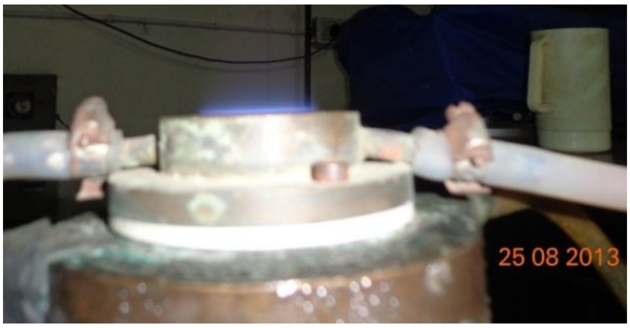

Fig.3. Direct photograph of stoichiometric Hythane (20% H2-80% CH4)-Air mixture at p=1 bar and T=292 K

Initially the experiments were started on the experimental set up developed by Yadav[12] at IIT Delhi and the set up was validated for CH4-Air stoichiometric mixture at 1 ba r and 292 and 298 K. Figure 5 shows the results of validation experiments. The obtained results are in we ll agreement with the results available in the liter ature within experimental uncertainty limits.

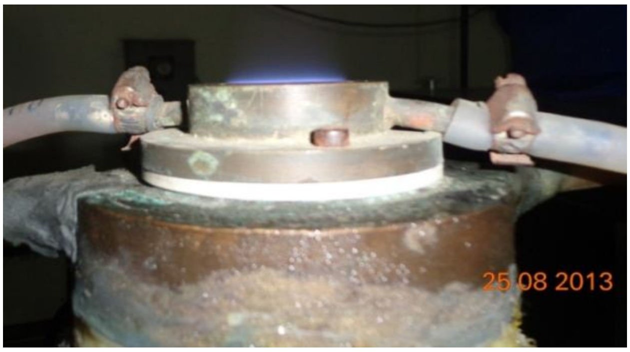

Fig.4. Direct photograph of stoichiometric Hythane (30% H2-70% CH4)-Air mixture at p=1 bar and T=292 K

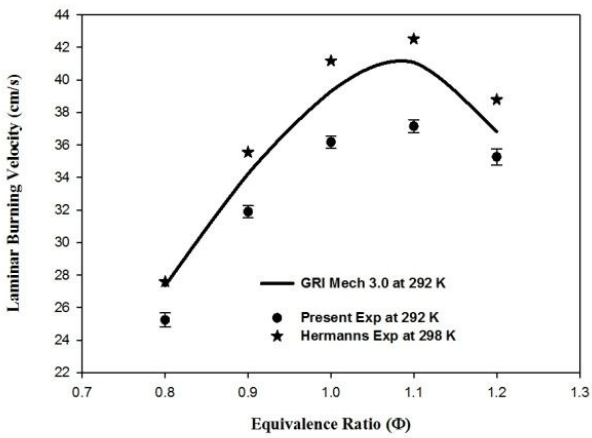

Fig.6. Experimental and computed laminar burning velocities (in cm/s) of 90% CH4 + 10% H2-Air at p = 1 bar, T = 292 K, Symbols: data; Lines: Numerical results

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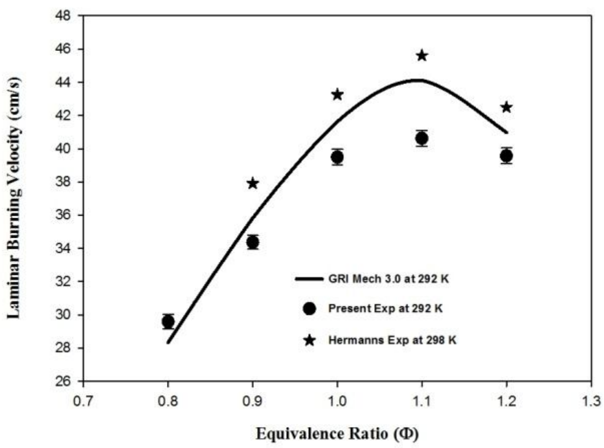

Fig.7. Experimental and computed laminar burning velocities(in cm/s) of 80% CH? + 20% H? at p = 1 bar, T = 292 K. Symbols:experimental data; Lines: numerical results

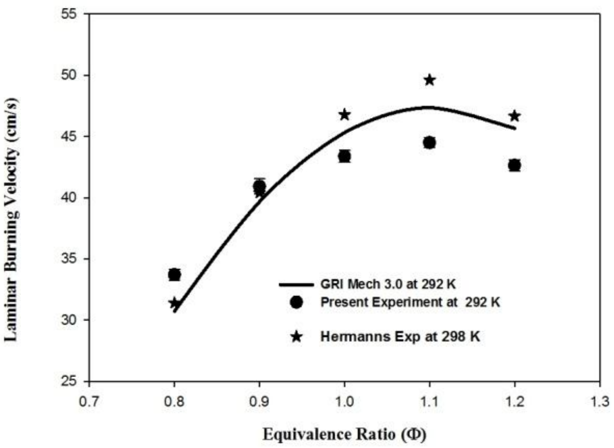

Fig.8. Experimental and computed laminar burning velocities (in cm/s) of 70% CH? + 30% H? at p = 1 bar, T = 292 K. Symbols: experimental data; Lines: numerical results

The experiments have been conducted with 10, 20 and 30 % H_2 enrichment in methane and the results are plotted in figure 6, 7 and 8 respectively. In all the cases, the predictions of GRI Mech 3.0 is in match with the present experimental results for lean and stoichiometric mixtures while the computational results are over predicting the LBV va lues in richer mixture side. The possible reason for the computational over prediction may be attributed t o the fact that GRI Mech 3.0 is best suited for CH_4-Air mixtures even at low temperatures. So, some other appropriate reaction mechanism may be used for the computational simulations. Hermanns [13] have conducted experiments at 298 K. So, the experimental results of Hermanns are higher than th e experimental results of the present work which is c arried at 292 K. It has been predicted that 4^0C increase in temperature increases the LBV by roughly 1 cm/s.

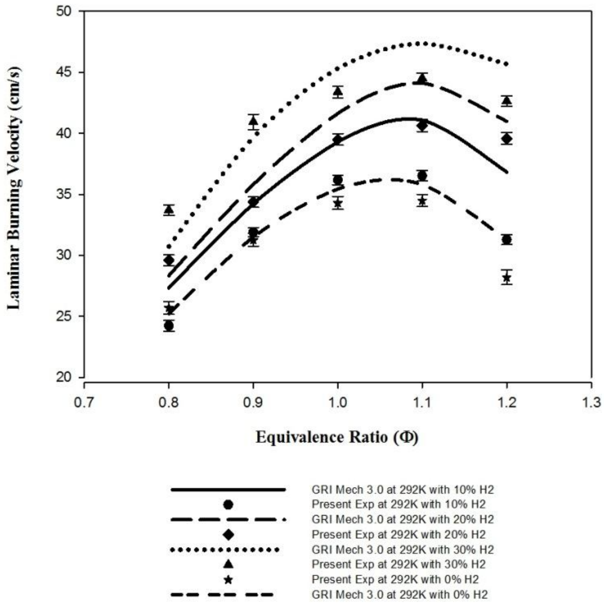

Fig.9. Unstrechted laminar burning velocity versus equivalence ratio at various hydrogen fractions at p = 1 bar, T = 292 K.

The combined experimental resu lts for 10, 20 and 30 % H_2 in methane and the results are plotted in figure 9. Correlation have bee n developed between laminar burning velocity and e quivalence ratio for 10% H_2 enrichment (Equation 1), 20% H_2 enrichment (Equation 2) and 30% H_2 enrichment (Equation 3).

$$S_L = -212.43\check{S}^2 + 249.65\,\check{S} - 194.86 \quad (1)$$

$$S_L = -111.71\check{S}^2 + 249.65\,\check{S} - 98.964 \quad (2)$$

$$S_L = -139.21\check{S}^2 + 299.86\,\check{S} - 116.83 \quad (3)$$

It has been observed that the peak LBV shifts towards richer mixture side with addition of H_2. It can be observed from Fig. 9 t hat for 0% H_2 in methane (i.e. pure CH_4), the peak LBV is behind equi valence ratio 1.1 while the peak is sh ifting towards right richer mixture as the hydrogen enrichment is increased. Further, it has been observed that with 10%, 20% and 30% H_2 enrichment the CH_4-Air LBV increased by 4.7 %, 15% and 22.47% respectively. This indicates an obvious fact of improvement in LBV of fuel mixtures with low heating values, which motivates to add the hydrogen with the gases like biogas and producer gas.

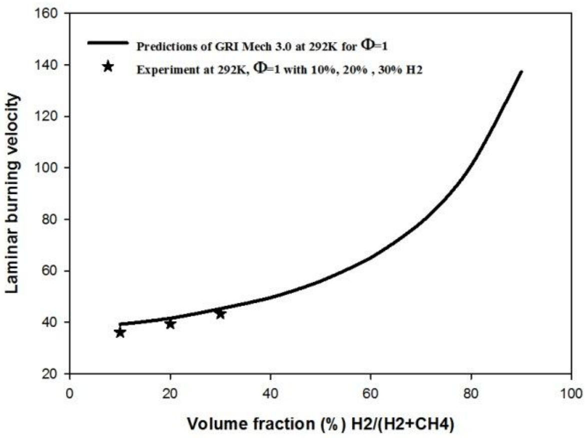

Fig.10. Unstretched burning velocity increment versus hydrogen fraction with stoichiometric HY1, HY2 and HY3 fuels at p=1 bar and T=292 K Symbols: experimental data; Lines: numerical results

Computations have been carried with GRI Mech 3.0 [10] by varying the H_2 fraction from 0 to 100 % in steps of 10 % and a correlation for the computational results is developed (Equation 4). So far, experiments have practically been conducted on the experimental set up for 10, 20 and 30 % H_2 enrichment. It can be observed from Fig 10 that the experimental results are following the trend of the computational results. Hence, on the available computational results a correlation between laminar burning velocity and hydrogen volume fraction (H_f) can be developed as

$$S_L = 0.0028\,H_f^2 + 0.248\,H_f + 33.42 \qquad (4)$$

It has been observed that by increasing the hydrogen content in methane the laminar burning velocity follows a trend of second order polynomial. Also there is a rapid rise in LBV after 60 to 70 % H_2 enrichment.

6. CONCLUSIONS

An experimental and computational study was carried at 292 K and atmospheric pressure on heat flux set up and following observations have been made:

- The Laminar Burning velocity of methane increases abruptly when it is blended with increasing percentage of hydrogen even at lower temperatures. A correlation has been developed to find out the expected LBV of various compositions of Hythane mixtures.
- The predictions of GRI Mech 3. 0 is in match with the present experimental results for lean and stoichiometric mixtures while the computational results are over predicting the LBV values in richer mixture side.
- The peak Laminar Burning Velocity of the mixture shifts towards richer mixture side with addition of H_2. A further shift towards richer mixture side may be observed by adding more hydrogen to the mixture.
- With 10%, 20% and 30% H_2 enrichment the CH_4-Air LBV increased by 4.7 %, 15% and 22.47% respectively. The computational trend predicts that if the hydrogen blending is increased further in the mixture, the laminar burning velocity will be improved further. The correlation between LBV and hydrogen fraction indicates that when hydrogen fraction is increased in the mixtures at lower temperatures, the LBV increment follows a second order polynomial trend.
- The temperature dependence of the laminar burning velocity can be well predicted . As a rough rule the experimental LBV increases by 1 cm/s for every 3 to 4 degree rise in temperature and vice-versa. In the present work, the experiments have been

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conducted at lower temperature s and the expected decrement in burning velocity is seen.

- The experimental results provided an idea to improve the stability, heating value and lean flammability limits of low hea ting value fuels like biogas, gasification gas and producer gas by blending them with hydrogen. F urther, the addition of hydrogen to conventional an d unconventional fuel improves their emission c haracteristics, which makes them suitable for enviro nment.
- The fuels having lower heating values when blended with suitable fraction of hydrogen may replace the conventional engin e fuels like petrol, diesel and CNG in gas turbines , household burners and internal combustion engine s.

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