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Inspection of Impurity effects on the methane laminar flame speed under lean and diluted conditions

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ABSTRACT

In the present work, the experimental study was performed to investigate the flame propagation and combustion characteristics of methane (CH_4)-air mixture in a constant-volume combustion vessel (CV) under lean ($\phi = 0.7$) and diluted engine-like condition. In addition, propane (C_3H_8) in two volume fractions of 20% and 40% was added to methane as the impurity and EGR gas. Schlieren photography of propagating spherical flame using a high speed camera and edge finding of flame pictures via a Matlab code was used to find the rate of flame instantaneous radius. The unstretched flame propagation speed and Markstein lengths were obtained via a nonlinear methodology in different chamber pressures. Flame speed modeling was performed using an improved version of kinetics model. The result showed that adding propane to methane increases the laminar flame speed and propensity of instability. The cellular instabilities for the diluted methane/propane–air flames were interpreted and evaluated in the viewpoint of the hydrodynamic and diffusional-thermal instabilities.

1) Introduction

There exist a large amount of methane-based gases in the earth, such as natural gas, landfill gas, and shale gas. use of gaseous fuels provides alternative energy resources and brings in safety and environmental benefits which are mentioned in many studies [1].

natural gas fuel in IC (Internal Combustion) engines has gained more attentions due to the increased oil prices and enforcement of stringent emission limits. Implementation of CNG (compressed natural gas) in spark ignition turbocharged engine offers high knocking resistance, low CO_2 emissions, and high specific power outputs [2].

CNG lean combustion is typically believed as a timely solution to the more stringent environmental regulations and global weather concerns. It is employed in nearly all industrial combustion systems, including gas turbine, boilers, furnaces, and internal combustion engines.

Stable combustion of CNG in lean conditions (air excess above 45 per cent) makes it possible to use the natural gas in SI engine for cogeneration (simultaneous production of electricity and heat) [3]. Stable combustion at ultra-lean conditions, makes it possible for CNG-fueled engines to operate with very low amount of emissions. In addition, lean burn also improves engine thermal efficiency by improving combustion quality, reducing heat transfer loss, and increasing possibility of applying higher compression ratios. NOx emissions are also reduced due to lower flame temperatures. Lean combustion of hydrocarbon fuels will result in complete burnout of fuel and reduces the hydrocarbon and carbon monoxide (CO) emissions.

However, slower flame propagation speed, increased cycle-by-cycle variations, and instability of combustion process are some difficulties of lean burn operation. These problems are due to the limited reaction rate, flame extinction, flame instabilities, mild heat release and sensitivity to mixing of natural gas [4]. Limited literature on concerns about the lean combustion in gas engines including ignition delay, combustion duration and combustion rate is available [5].

Rivin et al. [6] investigated the extension for lean misfire limit of methane-air mixtures by means of an enhanced-spark discharge at various operating conditions. Yoon et al. [7] did experiments to analyze the combustion characteristics of lean methane-air mixture. Their experiments were done on a wide range of initial pressures and turbulence flow conditions. They concluded that the ignition delay and combustion duration were increased by increase of the mixture pressure. The flame propagation speed has the highest value at the early stage of combustion and then will be decreased toward the end of combustion. The flame kernel growth of methane–air mixture at the engine-like conditions was done by Zhang et al. experimentally [8]. In their work, the flame kernel propagation and global combustion performances in a constant-volume chamber has been reported.

Usually, the flame conditions during the early flame development are nearly laminar due to small size of the flame compared to the integral length of turbulence.

Laminar burning velocity is an important parameter in combustion as it contains fundamental information regarding the reactivity and diffusivity. It is used in engine combustion simulations to predict turbulent burning velocity which is related to burn duration and It can be also used to validate chemical reaction mechanisms.

Many researchers have investigated the effects of dilution on laminar burning velocity of methane [9-12]. Numerical efforts have been done by Chemkin package or computer codes based on the detailed kinetic mechanism GRI-Mech 3.0. CO_2, H_2O and nitrogen (N_2) were used as dilution gases to represent the exhaust gases issued from the combustion of methane. They showed that the carbon dioxide dilution has a much larger suppression effect on decreasing the laminar burning velocity than nitrogen. For a similar flame speed in wet conditions, a further reduction of the burning velocity for N2 dilution than CO2 was reported.

It is also notable that the natural gas composition varies with location and season. In fact, the methane composition of natural gas can vary from 55.8% to 98.1%, ethane can vary from 0.5% to 13.3%, and propane can vary from 0% to 23.7% [5].

Different fuel composition result in significant operation problems for gas turbines and SI engines because of the variation of the flame speed [5]. Laminar flame speed of fuel blends which contain natural gas or methane concerned in recent years. Chen et al. [13] performed theoretical analysis and presented a model for the laminar flame speed of binary fuel blends. Their model showed that the laminar flame speed of binary fuel blends depends on the square of the laminar flame speed of each individual fuel component. Baloo et al. [14, 15] investigated the effect of methane addition to isooctane to find the effect of temperature and pressure on laminar burning velocities and flame instability of pure and blended fuels. They showed, methane addition to isooctane leads to more stable flame front. Lowry et al. [16] investigated the laminar flame speed of methane-dimethyl ether fuel blends. They also measured the flame stability, Markstein length, and Lewis number.

The combustion properties of the methane/propane blend have been reported in few publications. Laminar flame speed of propane as the main part of LPG and landfill gas was reported in the previous works [17-20]. Due to the impurity of landfill gas and LPG, the laminar flame speed of propane/methane blends with the specified percentage of each gas has

not been addressed in the literature. In the present study outwardly propagating spherical laminar premixed flames are investigated. The sensitivities of laminar burning velocities to the flame stretch; represented by Markstein length and Lewis number is also expressed. Propane (C3H8) in two volume fractions of 20% and 40% is also added to the methane (CH4) in order to investigate the effect of impurity on laminar burning velocity. Furthermore, the effect of EGR (exhaust gas recirculation) on burning velocities and instability of flame is investigated with adding 20% (in mass) N2 and CO2 to all mixtures as the EGR gas.

2) Experimental Approach

2-1) Facility

The experimental setup is shown in Fig. 1 and consists of a constant-volume combustion vessel (CV) with a volume of 1.1L, a fuel supply system, an ignition box and a data acquisition system in Michigan Tech Alternative Fuels Combustion Laboratory (AFCL). The temporal evolution of expanding flame was recorded via a high-speed schlieren photography.

Mixtures were prepared in a separate mixing vessel with partial pressure method. The CV is designed for a pressure up to 345 bar and the temperature up to 2000 K. The internal pressure of the vessel was continuously monitored via a piezo absolute pressure transducer (Kistler, 6001) connected to a charge amplifier (Kistler, 5044a). A spark plug holder is mounted in the CV top window. The electrodes of spark plug are extended to the center of chamber by welding of two steel rods (see Fig. 2). The spark gap was fixed to 1.3 mm. The pressure trace was recorded using a high-speed data acquisition system synchronized with the ignition and high-speed imaging systems.

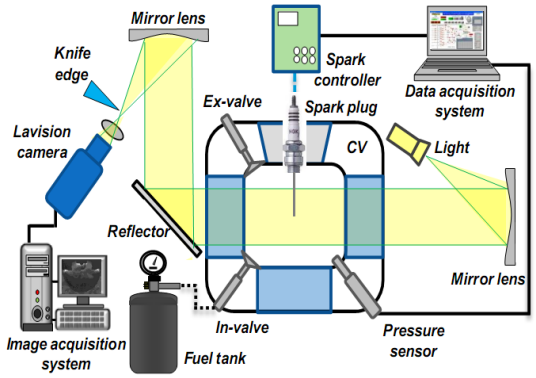

Fig. 1: A schematic of experimental combustion vessel and high-speed optical system.

Fig. 2: Spark plug with extended electrodes.

2-2) Mixture Preparation

All tests were done at an initial temperature of 423 K and three initial pressures of 2, 5 and 10 bar. Each test was repeated at least 3 times to ensure the repeatability. Propane was added to methane in two volume fractions of 20 and 40 percent. Three different fuel/air mixtures at $\phi = 0.7$ with 20% of EGR (by mass-base) were prepared. Table 1 summarizes the mole fraction of reactants in three fuel/air mixtures. Blended fuel can be expressed as $mC_3H_8 + (1-m)CH_4$ where m is propane mole fraction in mixed fuel.

Table 1: Mole fraction of species in different fuel/air mixtures with 20% EGR (in mass)

% Propane	X_{Fuel}	X_{C3H8}	X_{CH4}	X_{CO2}	X_{O2}	X_{N2}
0	0.057	0	0.057	0.014	0.166	0.766
20	0.044	0.009	0.035	0.015	0.168	0.774
40	0.036	0.015	0.022	0.016	0.170	0.780

2-3) Flame front Analysis

Flame front were traced by in-house developed MATLAB code. In order to remove the local disturbances and waviness, data is processed using the MATLAB® curve fitting tool. RLOESS (robust local regression using weighted linear least squares and a 2nd degree polynomial model) algorithm is then applied with a window size of 0.95 [14].

Fig. 3 shows the flame growth as a function of time for different mixture at the initial pressure of 5 bar and

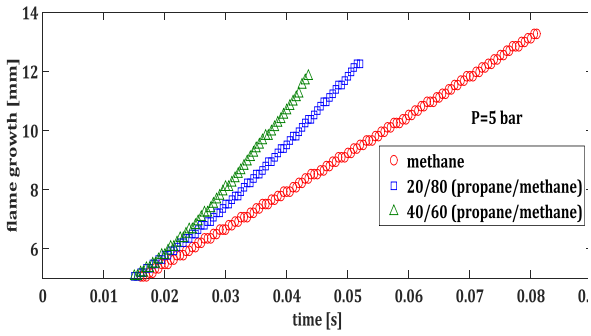

Fig. 3: Flame growth as a function of time for different mixture at P_{ini}= 5 bar.

Fig. 4 demonstrate the flame growth at different pressure for methane/air mixture with 20% EGR. As shown in Fig. 3, increasing the percentage of propane would result in the faster flame growth. It is also notable that the gradient of flame growth from 40% propane is more than that of 20% propane and pure methane. The flame growth decreases at higher chamber pressure due to the counterpoise of chamber pressure on the flame growth.

In order to avoid the effects of spark ignition disturbance [21-23] and the wall confinement [24] on flame propagation speed calculation, images with the radius range between 5 mm and 15 mm were selected to obtain the flame propagation

speed. Fig. 5 shows the pressure trace inside the CV during the test. In this selected range, data acquirement was performed at nearly constant chamber pressure ($\Delta p/p\_0 < 4\%$) during the initial stage of the flame expansion.

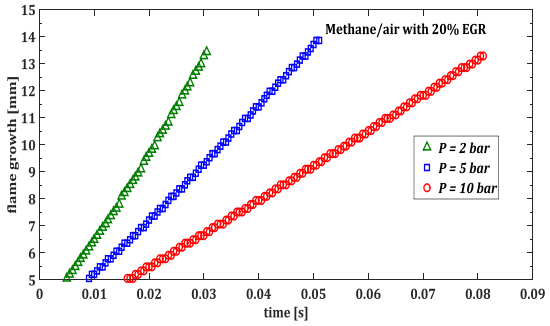

Fig. 4: Flame growth as a function of time for different pressure for methane/air mixture with 20%EGR.

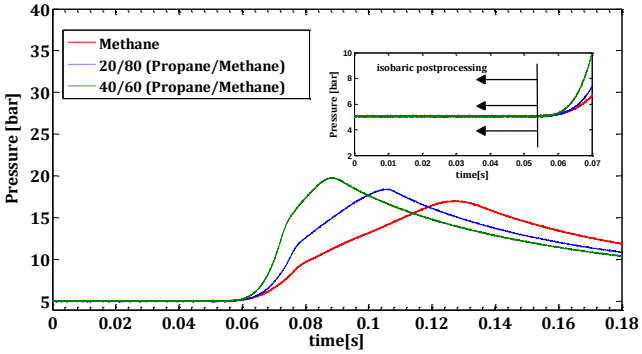

Fig. 5: Dynamic pressure in CV after combustion for three fuel/air mixtures.

In spherically outward expanding flame, the flame propagation speed can be calculated according to the temporal derivative of the flame radius growth:

$$S_b = \frac{dr}{dt} \quad (1)$$

Spherically growing flames are subjected to a variation of flame stretch that originates from the strain and the curvature of flame surface. The stretch rate is uniform during the constant pressure process and can be calculated as:

$$\alpha = \frac{1}{A}\frac{dA}{dt} = \frac{1}{4\pi r^2}\frac{d(4\pi r^2)}{dt} = \frac{2}{r}\frac{dr}{dt} \quad (2)$$

The unstretched flame propagation speed can be extracted as explained by linear method in Refs [25, 26]:

$$S_b = S_b^0 - L_b\alpha \quad (3)$$

where S_b^0 and S_b are the unstretched and stretched flame propagation speed, respectively, L_b is the Markstein length relative to burned gas, and α is the flame stretch rate. Markstein length is the representative of the flame response to the stretch rate.

S_b^0 is extracted from the linear extrapolation based on the plot of $S_b - \alpha$ as the intercept value of S_b at $\alpha = 0$. The slope of this line gives L_b. However, this linear relation is the result of several assumptions [27].

By considering nonlinear relations for the propagation speed of stretched flame, Kelley and Law [28] gave the nonlinear equation for the propagation speed as follows:

$$\left(\frac{S_b}{S_b^0}\right)^2 \ln\left(\frac{S_b}{S_b^0}\right)^2 = -2\frac{L_b \propto}{S_b^0} \quad (4)$$

In this paper, nonlinear method is used for assessing the flame propagation speed and Markstein length.

Finally, considering the mass conservation over the flame front with the assumption of quasi-steady and quasi-planar flame [29], unstretched laminar burning velocity S_u^0 is obtained:

$$S_u^0 = \frac{\rho_b}{\rho_u} S_b^0 \quad (5)$$

whereas ρ_b and ρ_u are burned and unburned gas density, respectively. The ratio ρ_u/ρ_b is the density of the burned gas, evaluated by chemical equilibrium computations using CHEMKIN at constant pressure, over the density of the reactants at the initial state. Normally this ratio is in the order of 0.25 for the mixtures studied in this work.

3) Results and Discussion

3-1) Laminar Burning Velocity

The molecular transport processes combined with the chemical kinetics determine the unstretched laminar burning velocity. This is the velocity at which the flame front propagates normal to itself, into the unburned mixture.

Laminar burning velocity extractions over the stretch rate of three fuel/air mixtures at initial pressure of 5 bar and 10 bar are shown in Figures 6 and 7. As shown in these figures, stretch decreases during the temporal evolution of the flame. The markers in the figures indicate the results of the post-processed images, and the lines represent the non-linear extrapolation of flame development according to Eq. (9). It is clear from, Figures 6 and 7 that with increasing initial pressure the stretched burning velocity is decreased.

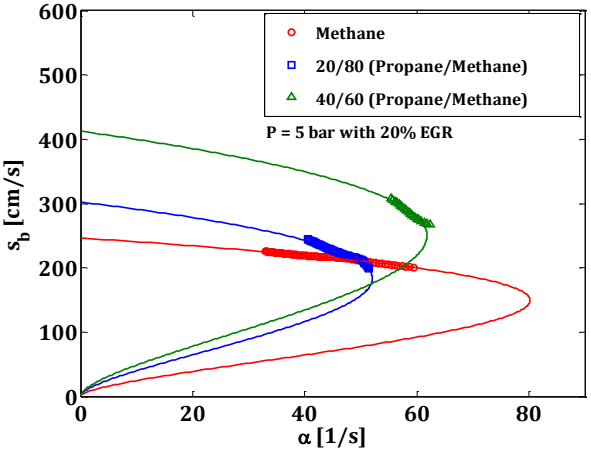

Fig.6: Propagation speed over stretch rate for three fuel/air mixtures with 20% EGR at P = 5 bar and T = 423 K.

At high pressures, the recombination reaction $H + O_2 + M \rightarrow HO_2 + M$ reduces the H atom concentration and thus competes with the initiation reaction producing free radicals O and OH: $H + O_2 \rightarrow O + OH$. Recombination reaction is third order and therefore much more pressure dependent than initiation reaction and this process tends to reduce the overall oxidation rate. Therefore, the combustion reaction is inhibited and the flame velocity decreases [30]. Also it is seen that the stretched burning velocity increases by adding the propane to methane.

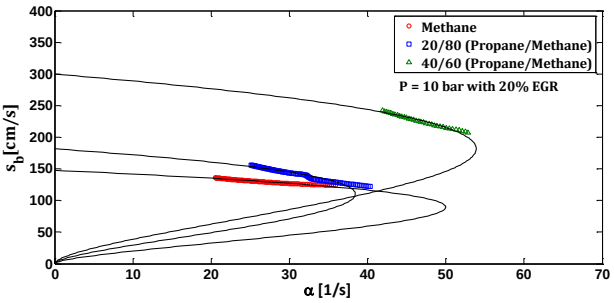

Fig.7. Propagation speed over stretch rate for three fuel/air mixtures with 20% EGR at P = 10 bar and T = 423 K.

3-1) Flame Instability

3-1-1) Markstein Length

The sensitivity of the flame propagation to the stretch rate is characterized by the Markstein length of burned gas, L_b. It can be either positive or negative depending on the reactant mixture. The positive value represents the flame is stable which means the flame surface is smooth and wrinkle-free. During the pre-pressure period of flame propagation, flame is stable toward the preferential-diffusion effects. The negative Markstein length indicates that the flame surface can become distorted due to preferential diffusion instability during its propagation, leading to the local acceleration of the flame and the formation of cellular structure. Small Markstein length indicates that the stretch does not have a considerable effect on the flame behavior.

3-1-2) Lewis Number Calculation

Lewis number (Le) has magnificent importance for the characterization of flame structure in gaseous fuels, particularly in the field of premixed combustion. The reaction front of premixed flames involves many chain reactions that are sustained by thermal conduction and species diffusion at leading edge, in preheat zone. Unequal heat and species diffusion could strongly affect the combustion intensity in the presence of flame stretch (flow straining, flame curvature).

Lewis number of a single fuel "i" in the mixture, was expressed as:

$$Le_i = \frac{D_T}{D_K} \quad (6)$$

where D_T is the thermal diffusivity of the fuel species "i", and D_k is the mass diffusion.

Mixture thermal diffusivity was calculated as:

$$D_T = \frac{\lambda}{\rho_u Cp} \quad (7)$$

Where ρ_u is the unburned mixture density, λ and C_p are the mixture-averaged thermal conductivity and specific heat, respectively.

λ for the multispecies mixture was determined by the following Mathur et al's recommendation [31].

$$\lambda = \frac{1}{2}\left(\sum_{k=1}^{N} X_k \lambda_k + \frac{1}{\sum_{k=1}^{N} \frac{X_k}{\lambda_k}}\right) \quad (8)$$

N being the total number of species in the mixture, X_k and λ_k the mole fraction and thermal conductivity of the k_{th} species, respectively.

C_p was also evaluated using the following mass fraction-weighted formulation:

$$C_p = \sum_{k=1}^{N} C_{pk} Y_k \quad (9)$$

where Y_k and Cp_k are the mass fraction and specific heat of the k_{th} species, respectively.

Multispecies diffusion is replaced by Hirschfelder and Curtiss approximation [32] which is the best first-order approximation to the exact resolution of diffusion velocities equation.

$$D_K = \frac{1-Y_k}{\sum_{i \neq k} X_i / D_{ik}} \quad (10)$$

Finally, the mixture Lewis numbers Le_{mix} were calculated using the effective formulation as proposed by Law et al. [33]

$$Le_{mix} = 1 + \frac{q_{CH4}(Le_{CH4}-1)+q_{C3H8}(Le_{C3H8}-1)}{q_{CH4}+q_{C3H8}} \quad (11)$$

where the parameter q_{C3H8} and q_{CH4} are the non-dimensional heat release associated with consumption of species C_3H_8 and CH_4 respectively and defined as:

$$q_i = \frac{QY_i}{CpT_u} \quad (12)$$

Here, Q is the heat of reaction and T_u is unburned gas temperature and Y is supply mass fraction (i=C_3H_8 or CH_4).

Table 2 shows that for blended fuels, Le_{mix} increases by adding more propane to methane. The results indicate that the flame tends to be unstable with increasing propane fraction. Markstein length and Lewis number increases with increasing the propane fractions, therefore the tendency of diffusive-thermal instability is enhanced.

Table 2. Lewis number, unstretched burning velocity and Markstein length for three fuel/air mixture with 20% EGR at p=5bar

Fuel	Le_{CH4}	Le_{C3H8}	Le_{mix}	S_u^0 cm/s	L_b mm
CH4	0.758	1.25	0.758	6.1	0.56
20/80 (C3H8/CH4)	0.753	1.22	0.944	7.3	0.87
40/60 (C3H8/CH4)	0.750	1.21	1.044	9.5	1.2

3-1-2) Buoyant Instability

The body-force instability was detected during the flame propagation in the experiments. The flame surface is always smooth, but the geometry of the flame is uplifted rather than maintaining the initial spherical shape during propagation. This phenomenon is pronounced at higher initial chamber pressure. The body-force instability has a repressive effect on the generation of irregular cracks on the flame front surface. The uncertainty associated with measuring the laminar burning velocity for lean mixtures with 20% EGR is almost due to the effect of buoyancy [34].

3-2) Numerical Simulation

PREMIX code [35] which is based on freely-propagating premixed planar flame with CHEMKIN package was used for prediction of unstretched laminar burning velocities. The burning velocity is defined as the linear velocity that the flame front travels relative to the unburned gas, and usually expressed as the volume of unburned gas consumed per unit time divided by the area of the flame front in a one-dimensional domain.

In the freely-propagating laminar one-dimensional flame, the mixing of fuel and oxidizer proceeds in the form of molecular diffusion, and it is expected that the flow rate of the mixture does not effect on the flame structure. As a result, the unstretched laminar burning velocity can be acquired.

The governing equations for isobaric, one-dimensional flame propagation are the continuity, conservation of, energy, species and equation of state. Mixture-averaged transport approximation and Soret effect were included in the computations. The Wang et al. [36] (USC Mech II) chemical kinetic scheme composed of 111 species and 784 reactions along with the corresponding transport and thermodynamic property data, was used in the simulations.

Unburned Initial temperature and the mass flow rate are 423 K and 0.04 gr/cm^{-2}s^{-1}, respectively. Fig.8 shows the comparison between the computed laminar burning velocities by using USC Mech II and experimental results. There is a good agreement found over a wide range of pressure. The results show that USC Mech II can well predict the laminar burning velocity of CH4/air flames at fuel-lean condition. However, for higher pressures, a better agreement is found with our experimental data and the combustion mechanism of USC Mech II.

4)Conclusions

This paper presents experimental results for binary blends of C_3H_8 and CH_4 with 20% EGR for two volumetric fractions performed at the initial temperature of 423 K and pressure of 2, 5, and 10 bar at the equivalence ration of $\emptyset = 0.7$ in the constant volume combustion vessel. Outwardly propagating spherical laminar premixed flames were studied to find the sensitive analysis of laminar burning velocities to flame stretch, represented by Markstein length and Lewis number.

The following conclusions are extracted from the results:

- The unstretched laminar burning velocities increase with higher propane fractions in methane while decreases with increasing the initial chamber pressure.
- The values of Markstein length and Lewis number increases with increasing the propane fractions, and therefore the tendency of diffusive-thermal instability is enhanced.
- The flame propagation speed for lean and diluted fuel/air mixture was too low ($S_u^0 < 16\ \frac{cm}{s}$) and therefore the body-force (buoyant) instability was detected during the flame move upward during propagation. This upward movement is more obvious in higher initial pressure.

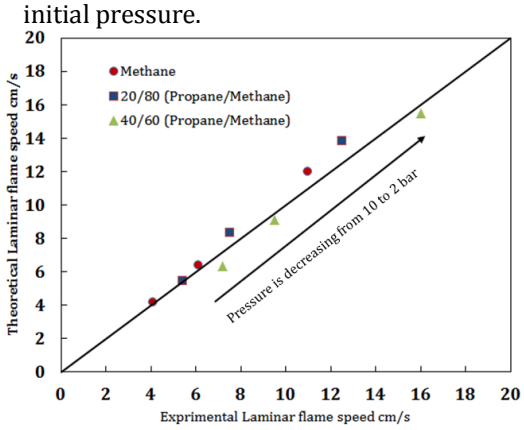

Fig.8: Comparison between the computed laminar flame speed and experimental results

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بررسی اثر ناخالصی سوخت در سرعت شعله آرام مخلوط رقیق متان

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چکیده

در این مقاله به مطالعه آزمایشگاهی سرعت شعله آرام و ویژگیهای احتراقی مخلوط متان و هوا در غنای ($\phi = 0.7$) و در شرایط موتوری با سوخت رقیق، در محفظه حجم ثابت پرداخته شده است. همچنین گاز پروپان با نسبت حجمی ۲۰٪ و ۴۰٪ حجمی به عنوان ناخالصی و گازهای نیتروژن، اکسیژن و دی اکسید کربن نیز به عنوان رقیق کننده و شبیه ساز گازهای برگشتی در موتور به مخلوط سوخت و هوا اضافه شد. شعاع لحظه‌ای شعله کروی و سرعت شعله آرام با در نظر گرفتن اثرات کشیدگی شعله با استفاده از روش عکسبرداری شیلیرین و روش لبه یابی درنرم افزار متلب، اندازه‌گیری و محاسبه شد. مدلسازی عددی سرعت شعله نیز با نرم افزار کمکین و با استفاده از مکانیزم بهبود یافته سینتیکی انجام و تطابق خوبی با نتایج اندازه گیری سرعت شعله آرام آزمایشگاهی مشاهده شد. نتایج نشان دادکه افزایش پروپان به متان رقیق شده با دود برگشتی، باعث افزایش سرعت شعله آرام و تمایل به ناپایداری بیشتر شعله می‌شود. همچنین ناپایداریهای سلولی سطح شعله با دسته بندی ناپایداری هیدرو دینامیکی و نفوذ حرارتی مورد بحث قرار گرفته است.

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