Numerical Simulation of the Effect of CH₄/CO Concentration on Combustion Characteristics of Low Calorific Value Syngas

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ABSTRACT: The composition of low calorific value synthesis gas varies greatly depending on the raw material and processing technology, which makes the combustion extremely complicated. The three mechanisms of the GRI-Mech 3.0, Li-Model, and FFCM-Mech are used to numerically simulate CH₄/CO/H₂/N₂ air premixed combustion by using ANSYS CHEMKIN-PRO. The numerical simulation is the calculation of laminar flame velocity and adiabatic flame temperature at an initial temperature of 298 K, an equivalence ratio of 0.6−1.4, and an initial pressure of 0.1−0.5 MPa, discussing through thermodynamics and chemical kinetics. The formation of NO, H, and OH radicals by fuel composition was analyzed. The result shows that the concentrations of H, O, and OH radicals have a positive effect on laminar flame velocity. The combustion reaction of H₂ is higher than that of CH₄ and CO; with the increase of N₂ content, the priority is higher. The thermal diffusivity of flame under different equivalence ratios is affected by inert gas, which affects adiabatic combustion temperature and laminar combustion velocity. In thermal kinetics and chemical kinetics, CH₄ has more influence on combustion temperature than CO, while laminar flame velocity is relatively low. Under the change of initial pressure, the laminar combustion flux increases to the initial pressure and the laminar combustion velocity decreases to the increase in pressure. Reactions H + O₂ = O + OH, HO₂ + H = 2OH, and CH₃ + HO₂ = OH + CH₃O are mainly due to change in the concentration of O, H, and OH radicals.

1. INTRODUCTION

Nowadays, the shortage of fossil energy and air pollution are increasingly affecting the living environment of human beings. The world is advocating energy saving and sustainable development.¹ It is very important for us to promote the development of renewable energy and establish the energy structure with new energy and renewable energy as the main body in the energy conversion process. Biomass is considered to be one of the most promising renewable sources of energy² because it is rich in resources and has the ability to absorb naturally released carbon and maintain carbon neutrality.³ In the past, direct combustion was the most important way to use biomass, which was often inefficient and caused serious pollution.⁴ Most gasified biomass are mixtures of H₂, CO, CH₄, N₂, CO₂ and other minor species.⁵⁻⁷ Biomass is the third largest renewable energy source used for electricity (after hydraulic and wind energy).⁸ In order to meet the growing demand, low calorific value syngas has gradually come into our view. In recent years, a large number of low calorific value natural gas are used in industries and homes. Since low calorific value syngas and conventional fuel exhibit diverse combustion features, the flame features will vary significantly, and with the increase of calorific value, flame instability may become a problem.¹⁰

For a long time, a large number of scholars have done a lot of research on hydrocarbon fuels. Biogas, landfill gas, and synthetic gases have also been studied due to the need for fossil energy substitution.¹¹ The methane content of biogas and landfill gas is about 40−60%, and the calorific value is about one-half of the calorific value of CH₄, which may be used as an alternative energy source.¹² In addition, the synthesis gas containing H₂/CO composition has been confirmed in which the flame stability will decrease as the CO ratio increases.¹³ Numerical simulations and experiments are used to verify the effect of adding diluent gases, such as N₂, He, and CO₂ in hydrocarbon fuels on flame stability.¹⁴ The addition of nitrogen exceeds about 10%, which is hard to demonstrate the flame stability.¹⁵ The laminar flame velocity of H₂/CH₄ and air plays an important role in explaining the combustion properties of combustible fuels, and it determines the stability of flame combustion and flame burning rate. Plenty of previous studies are about the laminar flame velocity of H₂/air mixtures,¹⁶⁻¹⁸ laminar burning rate of CH₄/air mixtures,¹⁹⁻²² laminar flame velocity of H₂/CH₄/air mixtures,²³⁻²⁶ and laminar flame...
velocity of CO/CH$_4$/H$_2$/air mixtures. Nowadays, there are a few studies on the laminar flame velocity of CH$_4$/CO/H$_2$/N$_2$ and air mixture.

For the study of the combustion features of low heating value gases (LHVGs), Shin et al. studied the flame stability restriction and flame length through a coaxial non-premixed jet burner. When the heating value is reduced, the flame stability limit value is significantly reduced. Francisco and Oliveira did not expect the effect of coaxial airflow; instead, the heating value of the fuel was reduced by increasing the nitrogen content in methane, and the flame structure was studied.

The purpose of this paper is to analyze the different combustion characteristics of CH$_4$/CO/H$_2$/N$_2$ air premixed flame by using ANSYS CHEMKIN-PRO; Table 1 details the composition of the fuel mixture; the laminar combustion velocity and adiabatic temperature under different equivalence ratios (0.6–1.4) were simulated by using the GRI-Mech 3.0, Li-Model, and FFCM-Mech mechanisms; and the results were discussed and analyzed. The effects of fuel components on the flame structure were discussed from the view of chemical kinetics, including the mole fraction of elements, yield, and net reaction rate of main reactions. The influence of initial pressure and fuel form of laminar combustion features were studied by sensitivity analysis of the initial pressure.

### 2. NUMERICAL CALCULATION

The characteristics of CH$_4$/CO/H$_2$/N$_2$/air laminar flame were calculated by CHEMKIN-PRO. One-dimensional, premixed, laminar flame model with the PREMIX code and gas-phase equilibrium program EQUIL were used to calculate the laminar flame propagation velocity and adiabatic temperature. In this calculation, three combustion kinetic mechanisms of GRI-Mech 3.0, Li-Model, and FFCM-Mech are used. The detailed reaction mechanism of the GRI-Mech 3.0 includes 325 reactions and 53 substances; the detailed reaction mechanism of Li-Model includes 84 reactions and 21 substances; and the detailed reaction mechanism of FFCM-Mech includes 291 reactions and 38 substances. Figure 1 shows a comparison with the previous literature, which shows that the results are in good agreement with different mechanisms. In view of the current calculation, the Soret effect and multicomponent transmission model are considered in the simulation. The calculation domain is set to −2 to 10 cm, and the GRAD and CURV values are set to 0.02 to meet the boundary conditions of the free flame propagation and no thermal diffusion. The number of grids is set to 900, which is used to determine the flame velocity prediction for complete convergence.

![Figure 1. Laminar flame speed of H$_2$ (40%)/CO (40%)/CH$_4$ (20%)/N$_2$/air mixtures.](https://dx.doi.org/10.1021/acsomega.0c06176)

### 3. RESULTS AND DISCUSSION

#### 3.1. Influence of Different Fuel Forms on Adiabatic Temperature and Laminar Combustion Speed

Figure 2 shows the variation distribution of premixed CH$_4$/CO/H$_2$/N$_2$/air adiabatic temperature and laminar combustion velocity under different fuel components simulated by CHEMKIN-PRO. The thermal diffusivity of different fuel components was calculated by STANJAN. It can be seen from Figure 2a that the thermal diffusivity of all simulated fuel mixtures increases monotonously at the equivalent ratio (0.6–1.4). As the proportion of CH$_4$ gas increases, the thermal diffusivity of the flame decreases significantly, while the influence of CO is significantly greater than CH$_4$; when the proportion of CO gas decreases, the thermal diffusion coefficient increases significantly. In conclusion, CH$_4$ fuel suppresses thermal diffusivity, and CO fuel increases thermal diffusivity. In addition, in Figure 2b, the adiabatic flame temperature increases significantly at the equivalent ratio of 0.6–1.0, reaches the peak at the equivalent ratio of 1.0, and decreases significantly at the equivalent ratio of 1.0–1.4. Compared with CO gas, CH$_4$ gas has a higher adiabatic flame temperature; the difference is about 200 K. Although the thermal diffusivity of the F6 fuel component is higher than the F1 fuel component, the adiabatic combustion temperature of the F6 fuel component is greater than that of the F1 fuel component. The combustion speed is slightly different due to the fuel composition. As shown in Figure 2c, the F3 fuel reaches the maximum when the equivalent ratio is 1.0, and the $S_L$ of the F4 flame increases significantly compared with the F3 flame. Second, the peak value of $S_L$ will move to a rich state with the increase of CO content, and the laminar flame velocity significantly increases compared with the F3 fuel composition condition. From the laminar flame theory, $S_L \propto (RR)^{1/2}$, the laminar flame velocity is positively related to the adiabatic flame temperature ($T_a$) and thermal diffusivity ($\alpha$) and is directly related to the reaction rate (RR). In addition, it is worthy of our attention that the three mechanisms of the GRI-Mech 3.0, Li-Model, and FFCM-Mech can predict the laminar flame velocity at different equivalence ratios. In the simulation of F6 fuel components, the peak velocity of the GRI-Mech 3.0 is more inclined to the fuel-rich part, which is slightly different from the other two mechanisms.

It can be known from previous papers that the effect of increasing the proportion of inert gas on the laminar combustion rate is mainly due to the increase in heat capacity and the decrease in flame temperature, thereby changing the
thermal performance of hydrocarbon fuels. The simulation results of this work are similar to the pure methane in the earlier papers. On the one hand, with the increase of the inert gas ratio, the concentration of fuel is diluted, resulting in the decrease of heat release and the reaction rate. Second, it will also affect the thermal diffusion coefficient and oxidation reaction kinetics of the mixture.

3.1.1. Flame Structures. Figure 3 shows the most important basic reaction for calculating the laminar combustion velocity of different fuel components (F3, F4, F6) using the GRI-Mech 3.0, Li-Model, and FFCM-Mech. The combustion occurred mainly in the range of 3.96–4.08 cm. Second, the combustion reaction of H2 is higher than that of CH4 and CO, and with the increase of N2 content, this priority is higher. It can be known in the GRI-Mech 3.0 that there are five main reactions where H2 affects the combustion speed, reactions O + H2 = H + OH (R3), H + O2 + H2O=HO2 + H2O (R35), H + O2 + N2 = HO2 + H2O (R36), H + O2 = O + OH (R38), and OH + CO=H + CO2 (R99). It can be learned from the Li-Model that H2 mainly has four important reactions affecting the combustion speed, reactions H + O2 = O + OH (R1), HO2 + H = 2OH (R15), CO + sOH=CO2 + H (R29), and CH3 + H(+M) = CH4(+M) (R53). In FFCM-Mech, it can be known that H2 affects the combustion rate mainly in four important reactions H+O 2 = O + OH (R1), H2O + M = H + OH + M (R15), CO + OH=H + CO2 (R32), and HCO + M = H + CO + M (R35). From these three mechanisms, we can know that reaction R1 is a branched chain reaction that generates a large amount of H and OH radicals. These active radicals (H, O, and OH) have an important effect on the laminar flame velocity.

3.1.1. Flame Structures. Figure 3 shows the most important basic reaction for calculating the laminar combustion velocity of different fuel components (F3, F4, F6) using the GRI-Mech 3.0, Li-Model, and FFCM-Mech. The combustion occurred mainly in the range of 3.96–4.08 cm. Second, the combustion reaction of H2 is higher than that of CH4 and CO, and with the increase of N2 content, this priority is higher. It can be known in the GRI-Mech 3.0 that there are five main reactions where H2 affects the combustion speed, reactions O + H2 = H + OH (R3), H + O2 + H2O=HO2 + H2O (R35), H + O2 + N2 = HO2 + H2O (R36), H + O2 = O + OH (R38), and OH + CO=H + CO2 (R99). It can be learned from the Li-Model that H2 mainly has four important reactions affecting the combustion speed, reactions H + O2 = O + OH (R1), HO2 + H = 2OH (R15), CO + sOH=CO2 + H (R29), and CH3 + H(+M) = CH4(+M) (R53). In FFCM-Mech, it can be known that H2 affects the combustion rate mainly in four important reactions H+O 2 = O + OH (R1), H2O + M = H + OH + M (R15), CO + OH=H + CO2 (R32), and HCO + M = H + CO + M (R35). From these three mechanisms, we can know that reaction R1 is a branched chain reaction that generates a large amount of H and OH radicals. These active radicals (H, O, and OH) have an important effect on the laminar flame velocity.

With the change of N2 in the fuel, when CH4 fuel is the main gas, H + O2 = O + OH (R38), it has the most important effect on combustion, and the H + CH3 (+M) = CH4 (+M) (R52) reaction has the highest negative sensitivity coefficient and inhibits flame propagation. At the same time, when CO is used as the main fuel, the OH + CO=H + CO2 (R99) reaction has the highest positive sensitivity coefficient, promoting the flame propagation. Meanwhile, reaction H + HO2 = O2 + H2 (R45) has a large negative sensitivity coefficient, reducing the flame propagation speed. When CH4/CO are used as the main fuel,
reaction $H + O_2 = O + OH$ (R38) has the maximum positive sensitivity coefficient.

3.1.2. Chemical Kinetic Structure. Three different fuel components F3, F4, and F6 were analyzed in detail by the GRI-Mech 3.0, Li-Model and FFCM-Mech mechanisms. Figures 4−6 show the mole fraction, productivity, and net reaction rate of the main basic steps, respectively. For F3 (20% CH$_4$, 10% H$_2$, 70% N$_2$), we can see that in the mechanism of...
using the GRI-Mech 3.0, Li-Model and FFCM-Mech, the consumption of CH₄ mole fraction is later than that of H₂, so the formation of H₂O is higher than that of any substances. The mole fraction of substances in F3 fuel shows similar change curves in the three mechanisms. In Figure 5a,d,g, we can know that H is consumed first and then generated, the consumption of CO is the latest, and the rate of CO₂ generation increases first and then decreases.

In the GRI-MECH 3.0, R38, R97, and R99 play a significant role in F3, generating a large number of O and OH radicals.
and CO$_2$. The removal of H from CH$_4$ mainly has the following reaction: H + CH$_4$ $\rightarrow$ CH$_3$ + H$_2$ (R53) and OH + CH$_4$ $\rightarrow$ CH$_3$ + H$_2$O (R98). Then, CO begins to oxidize, passing OH + CO $\rightarrow$ H + CO$_2$ (R99). The intermediate product CO is mainly composed of H + CH$_4$ $\rightarrow$ CH$_3$ + H$_2$ (R53) and O + CH$_3$ $\Rightarrow$ H + H$_2$ + CO (R284). At the same time, the CH$_3$ can also terminate the free radical chain process through H + CH$_3$ + M $\rightarrow$ CH$_4$ + M.

For the F4 fuel (10% CH$_4$, 10% CO, 10% H$_2$, 70% N$_2$), due to the addition of CO, we can see in Figure 4 that the CO
content increases slowly and then decreases, while for the F3 fuel, the CO content rises sharply, and then declines slowly. For the content of CH₃-free radicals, the content of the F3 fuel is significantly higher than that of the F4 fuel. In Figure 5b,e,h, we can see that the content of CO generated by the oxidation of CH₄ is greater than the consumption of CO. Obviously, the combustion of CH₄ precedes CO. In Figure 6e, in the Li-Model mechanism, R1, R3, R29, and R48 play a significant role in the F4 fuel, generating a large number of free radicals of O, H, OH, and CO₂. The reaction rate of R29 in the F4 fuel is significantly higher than that of the F3 fuel, thereby increasing the overall reaction rate. Therefore, the F4 fuel has a higher flame laminar flow velocity than the F3 fuel.

For the F6 fuel (20% CO, 10% H₂, 70% N₂), compared with the F3 fuel, CH₄ becomes CO. It is learned in Figure 4c,f,i that H₂O is generated later than the F3 fuel, and the content of CO₂ is higher than that of the F3 fuel. Compared with the F4 fuel, the F6 fuel consumes H₂ faster, and the content of H₂O is greater than that of CO₂. In Figure 6i, it can be found that compared with the previous two flames, the main chemical composition of the flame has been transferred to CO kinetics. R1, R4, R15, and R32 are the main reactions, generating a large number of free radicals of O, H, OH radicals and H₂O and CO₂ substances. The net rate of the chemical reaction is much lower than that of the F3 fuel and F4 fuel. From the previous results of the laminar combustion temperature in Figure 2b and the laminar combustion velocity in Figure 2c, it is found that the F6 fuel laminar combustion temperature and the laminar combustion speed are higher than that of the F3 fuel flame and the F4 fuel flame. This is the same as the conclusion of Vagelopoulou et al.⁴⁴ that the CO added into the fuel mixture will not work until lots of the hydrocarbons are consumed, and its effect is essentially heat.

3.1.3. Influence of Inert Gas on NOₓ Formation. NOₓ emissions have a great impact on air. On the one hand, NOₓ is formed by the nitrogen element in the fuel; on the other hand, it is mainly formed by the oxidation of N₂ in air. The effect of inert gas on NOₓ production in CH₄ combustion has been reported in the previous literature.⁴⁵,⁴⁶ Figure 7 shows the changes in the maximum mole fraction of NO and NO₂ for F3, F4, and F6 fuels through the GRI-Mech 3.0 mechanism. As shown in the figure, CO has an obvious inhibitory effect on the formation of NO, but the inhibition of NO₂ during the combustion process is not very obvious. However, CH₄ has a very obvious effect on the formation of NO and NO₂. Since the concentration of NO₂ is much lower than NO, the NOₓ in the whole reaction is related to the content of CH₄.

3.2. Impact of Pressure. Figure 8 reveals the impact of using the FFCM mechanism at different initial pressures (0.1, 0.3, and 0.5 MPa) and equivalence ratios on the combustion rate of F4 (10% CH₄, 10% CO, 10% H₂, 70% N₂) fuel laminar flow. We can know that the laminar combustion speed of the flame and the thermal diffusivity in the fuel mixture decrease with the increase of the initial pressure.⁵ Therefore, when the
equivalence ratio $\phi = 1.0$, there is a maximum laminar combustion velocity.

3.2.1. Laminar Combustion Flux. From the study of Law and Sung,\textsuperscript{47} the laminar combustion flux $f_0 = \rho_u S_b$ is the basic modulus of flame propagation. It generally reflects fuel diffusivity, exothermicity, and reactivity. Figure 9 shows the laminar combustion flux of F4 (10% CH$_4$, 10% CO, 10% H$_2$, 70% N$_2$) under different initial pressures and equivalence ratios. From the figure, we can find that the laminar combustion speed of F4 is the largest at 0.1 MPa; but on the contrary, it has a smaller laminar combustion flux. The laminar combustion flux of F4 improves with the increase of premier pressure, which is similar to Law’s\textsuperscript{48} conclusion. He pointed out that the laminar flow velocity of flame decreases with the increase of initial pressure due to the increase of density. According to the conclusion of Law et al.,\textsuperscript{47} we know that $S_{b0} \approx \left[\frac{\lambda}{c_p}\nu_w\right]^{1/2}/\rho_b$. This reveals that laminar flame responses rest with the flame dynamics of the typical reaction rate $\nu_w$ and the transport processes through the density-weighted transport parameter ($\lambda/c_p$)$_w$. For us, density is the most crucial factor, which determines the explanation of the character of diffuse transport the same as that of the volume mass flow rate.\textsuperscript{16}

3.2.2. Sensitivity Analysis. Figure 10 shows the important basic reactions that influence the combustion velocity of laminar flame at diverse premier pressures. With the increase of pressure, the number of molecular and free radical collisions is improved and the reaction rate is accelerated. In the FFCM-Mech, we can find that with the increase of initial pressure, the sensitivity of the reaction R1: H + O$_2$ = O + OH, R17: HO$_2$ + H = 2OH, R105: CH$_3$ + HO$_2$ = OH + CH$_3$O and the coefficient also increase accordingly. However, the negative sensitivity coefficient R15: H$_2$O + M = H + OH + M, R16: HO$_2$ + H = H$_2$ + O$_2$, R97: CH$_3$ + O ⇒ H + H$_2$ + CO also increased, which inhibited the propagation of the laminar flame velocity. In the previous literature,\textsuperscript{49} termination of the reaction was very important as the pressure increased. If we add H and OH radicals and reduce HO$_2$ radicals and a small amount of H$_2$O molecules, so far, it will promote the entire reaction, and the flame burning rate will be greatly increased.

3.2.3. Chemical Kinetic Analysis. According to previous research,\textsuperscript{50,51} it is known that the main influencing factors of the laminar flame velocity have a strong positive correlation with the concentration of free radicals H, OH, and O. Figure 11 shows the radical mole fraction of F4 fuels under the FFCM-Mech at different initial pressures. We found that with the increase of the initial pressure, the concentration of H, OH, and OH radicals decreased, which is basically similar to the change of the flame laminar combustion speed.

4. CONCLUSIONS

The laminar flame velocity of syngas with a low calorific value at an equivalence ratio of 0.6−1.4 and an initial pressure of 0.1−0.5 MPa was studied by using ANSYS-CHEMKIN, and the effects on thermal diffusivity, formation of NO$_x$, H, and
OH radicals were analyzed. The main conclusions are summarized as follows:

(i) Compared with CO, CH₄ has a larger adiabatic temperature and a smaller laminar flame velocity. Inert gas affects the development trend of flame thermal diffusion and reduces the adiabatic temperature and laminar combustion velocity.

(ii) The three mechanisms of GRI-Mech 3.0, Li-Model, and FFCM-Mech predict the laminar flame velocity at different equivalence ratios and show good consistency among them. In the preflame region, the peak velocity of the GRI-Mech 3.0 is more inclined to the fuel-rich part than that of the Li-Model and FFCM-Mech.

(iii) In the flame structure, the reaction of H₂ always takes precedence over CH₄ and CO. R1 is a branched reaction that produces a large number of O, H, and OH radicals, which have a positive effect on the laminar flame speed. When CO and CH₄ are the main fuels, it is found that there is the same elementary reaction as CH₄ as the main fuel. In the mixed gas, CH₄ gas plays a major role.

(iv) With the increase of the initial pressure, the laminar combustion speed of the flame and the thermal diffusion coefficient of the fuel mixture decrease accordingly. The increase in pressure increases the number of collisions of active free radicals and speeds up the reaction rate. If we add O, H, and OH radicals, it will promote the entire reaction.

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### Notes

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