

## LAMINAR PREMIXED BURNING CHARACTERISTICS OF METHANOL/ETHANOL/AIR AT HIGH TEMPERATURE AND PRESSURE

by

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*A numerical study was conducted to evaluate the laminar burning characteristics of methanol/ethanol/air flames under engine operating conditions. Methanol and ethanol are common burning oil of biomass that can replace fossil fuels and have a high laminar combustion rate relative to gasoline. Few scholars have studied the laminar combustion characteristics of methanol/ethanol mixture. This study aims to analyze the similarities and differences of methanol/ethanol/air flames for different methanol percentages and equivalence ratios in a wide range of pressure and temperature conditions. Chemkin software with chemical reaction kinetic mechanism of Mittal was used and validated against experimental data from the literature. The results showed that methanol significantly increases the laminar burning velocities and the net heat release rate. The maximum laminar burning velocities is obtained at the equivalence ratio of 1.1. Due to the high calorific value of ethanol, the adiabatic flame temperature of ethanol are slightly higher than that of methanol. The hydrodynamic instabilities of methanol/ethanol/air flame increase at first and then decrease with the increase of equivalence ratio. Finally, sensitivity analysis show that  $H + O_2 \rightleftharpoons O + OH$  (R1) mainly influences the laminar burning velocities.*

**Key words:** laminar burning velocity, methanol, ethanol, sensitive analysis

### Introduction

To achieve carbon peak and neutrality targets, the search for clean renewable alternative fuels has become an important area of scientific research. Methanol and ethanol have the characteristics of low carbon, efficient combustion, clean emission and renewable, and are liquid under normal atmospheric pressure and temperature, which is safer and more convenient to store, transport and use than other energy carriers [1, 2]. Methanol and ethanol have similar compatible properties and can be synthesized chemically and biologically. They have a high octane number and can be used efficiently in engines [3]. Furthermore, their clean-burning properties can reduce the emission of harmful gases [4].

To improve combustion efficiency and reduce pollutant emissions, scholars have conducted research on different fuel combinations under a wide range of engine operating

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conditions. Beeckmann *et al.* [5] carried out experiments on the laminar burning velocity of isooctane, methanol, ethanol, isooctane/methanol, and isooctane/ethanol, and conducted numerical simulations using high temperature chemical models. Vancoillie *et al.* [6] reviewed the published experimental data of laminar burning velocity of methanol-air and ethanol-air, and presented two laminar burning velocity correlations covering the entire working range of alcohol fuel spark-ignition engines. Brian *et al.* [7] have studied the spontaneous combustion performance of various alcohols in a homogeneous compression ignition and found that all kinds of alcohol fuels have similar sensitivity to various operating conditions, which means that common alcohol fuels may be mixed and used interchangeably. Sileghem *et al.* [8] studied the laminar burning velocity of methanol and ethanol as a function of equivalent ratio, and found three correlations that can be used in simulation models. Budi *et al.* [9] studied and tested the performance of stable homogeneous gasoline-methanol-ethanol blends in a single-cylinder engine. They found that less than 30% of the gasoline-methanol-ethanol blends produced more power than gasoline under all operating conditions because of its higher laminar burning velocity [10].

Scholars have conducted some experimental and numerical studies on the laminar burning velocity of methanol [11] or ethanol [2, 12]. However, few scholars have conducted chemical reaction kinetic simulation studies on the premixed laminar burning characteristics of methanol/ethanol blends [13]. Therefore, in this paper, the effect of various parameters on the laminar burning characteristics of methanol/ethanol/air flames is comprehensively analyzed by using 1-D numerical simulations. The kinetic characteristics of the combustion process of the mixed fuel are analyzed. The research results enrich the basic data of methanol-ethanol-air laminar premixed flame, and provide a basis for the design of methanol-ethanol mixed fuel engine.

## Method

### Methodology

In this paper, thermodynamic and kinetic simulations were performed with CHEMKIN software using Mittal's mechanism (consists of 113 species and 710 reversible reactions [14]). The LBV was calculated with the 1-D freely propagating premixed code. The physical model of laminar combustion was simplified into a mathematical model in CHEMKIN. The 1-D flow is assumed to have uniform entrance conditions. The governing conservation equation is simplified to:

- Continuing equations

$$\dot{m} = \rho u A \quad (1)$$

- Energy equations

$$\dot{m} \frac{dT}{dx} - \frac{1}{c_p} \left( \lambda A \frac{dT}{dx} \right) + \frac{A}{c_p} \sum_{k=1}^K \rho Y_k V_k c_{p,k} \frac{dT}{dx} + \frac{A}{c_p} \sum_{k=1}^K \dot{\omega}_k h_k W_k + \frac{A}{c_p} \dot{Q}_{\text{rad}} = 0 \quad (2)$$

- Species equations

$$\dot{m} \frac{dY_k}{dx} + \frac{d}{dx} (\rho A Y_k V_k) - A \dot{\omega}_k w_k = 0, \quad k = 1, 2, \dots, K \quad (3)$$

- State equations:

$$\rho = \frac{P \bar{W}}{RT} \quad (4)$$

These formulas were used to obtain the thermodynamic data and dynamic components, which were then used to calculate the laminar flame propagation speed [15]. Table 1 summarizes the fuel composition and operating conditions used in this paper (M70 represents a fuel mixture with a mole fraction of 70% and 30% of methanol and ethanol, respectively; the same for M50 and M30).

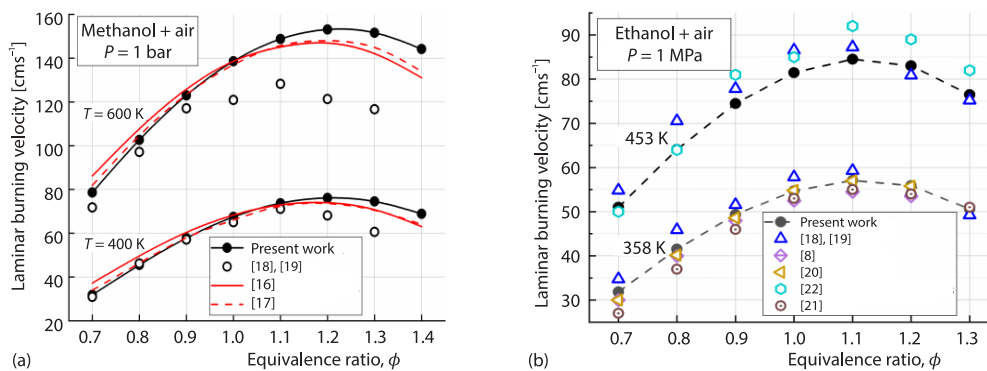
**Table 1. Fuel composition and working condition**

Fuel	Pressure [bar]	Temperature [K]	Equivalence ratio
Ethanol	1-40	400-800	0.7-1.4
M30	1-40	400-800	0.7-1.4
M50	1-40	400-800	0.7-1.4
M70	1-40	400-800	0.7-1.4
Methanol	1-40	400-800	0.7-1.4

The mixture-averaged formula was used in the simulations to determine the species' ordinary diffusion coefficients and fluxes. This is less accurate than the full multicomponent approach but is usually computationally faster and often easier to converge. To ensure that the calculated value is in the thermodynamic equilibrium state, that is, all the heat release of the reaction is used to increase the temperature, the starting point of the reactor calculation is set as 0 cm and the endpoint as 0.3 cm in the model. The curvature and gradient of the calculated curve converge to 0.1 and 0.05, respectively. The number of grids is greater than 250 and the maximum is less than 2000.

*Model validation*

Figure 1(a) shows the LBV comparison of methanol at 400 K and 600 K. The LBV simulated with Mittal's mechanism in this paper has the same tendency as the results obtained by San Diego Mech [16] and Li Mech 1 [17]. The Mittal's mechanism match well with the predictions of the two detailed kinetic models at mixture temperature of 600 K. The data measured by Katoch *et al.* [18] showed some deviation with the simulation values for the rich mixture, which can perhaps be attributed to the existence of uncertainty factors in the experiment, such as the heat loss. In the model, the system is assumed to be adiabatic, while in the experiment, adiabatic conditions cannot be achieved, so the simulation results are larger than the experimental ones.



**Figure 1. The LBV of methanol and ethanol was obtained in this work and the literature; (a) methanol and (b) ethanol**

In the case of ethanol, the LBV simulated with Mittal's mechanism in this paper agrees well with Katoch [19], Sileghem [8], Dirrenberger [20], and Konnov [21] at 358 K, as shown in fig. 1(b). The difference between Katoch [19] and Saxena and Williams [22] at 453 K might be due to the researchers' different experimental and postprocessing methods. For both 358 K and 453 K, the LBV of the present study has a similar tendency as that of the literature data.

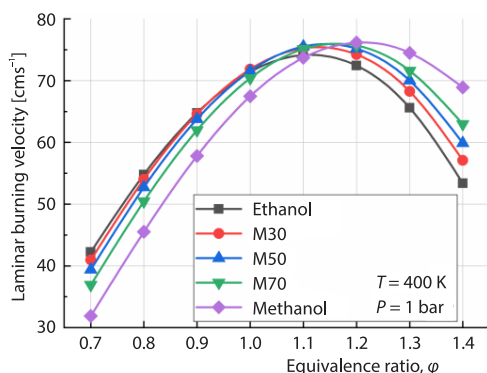
The numerical model is adiabatic but the experiment is non-adiabatic. The model reasonably predicts the variation trend of the LBV with the equivalence ratio under the adiabatic condition. Consequently, this research can use the model to reasonably predict the premixed laminar combustion characteristics of the mixed fuel.

## Result and discussion

### Laminar burning velocity

The LBV of methanol/ethanol/air flames at an initial temperature of 400 K and an initial pressure of 1 bar are shown in fig. 2. The LBV first increases and then decreases, and reaches the peak when the equivalence ratio is 1.1 except for the pure methanol fuel. At the same time, the LBV increases with the increase of methanol content when  $\varphi > 1.2$ .

The main reactions of alcohol fuel combustion are marked by the dehydrogenation reaction caused by the breakdown of the C-H bond [23]. The number of C-H bonds in different fuels is different, and the molecular collision energy between the reactants in the main reactions



**Figure 2.** The LBV of methanol/ethanol/air flames; data are given for 1 bar and 400 K

of alcohol fuel combustion is different, therefore, there are different laminar burning characteristics. Methanol's C-H bond dissociation energy reduces the laminar combustion rate, leading to a poor lean combustion limit. However, at  $\varphi > 1.2$ , the LBV of pure methanol is much higher than other mixed fuels, indicating that methanol has better combustion enrichment characteristics. In rarefied combustion, the LBV of the mixed fuel is similar except for methanol. At the same time, LBV reaches the maximum value when the equivalence ratio is 1.1. This is due to the proportion of intermediate combustion products with small LBV reacting with air at this relatively small equivalence ratio.

One of the most important factors affecting the LBV is the initial temperature. According to fig. 3, the LBV of methanol/ethanol/air flames increases with increasing temperature. Higher temperature increases the activity factor of the reactants, enhances the chain structure reaction, and speeds up the chemical reaction rate. The trend of methanol/ethanol/air flames LBVs with temperature is almost the same. The initial temperature affects the LBV in three ways [24]. Firstly, it influences the LBV by changing the reaction rate and flame temperature. Next, it changes the transport properties of the mixed fuel to affect the LBV. Finally, the LBV is affected by the change in density.

Figure 4 shows the variation of different fuels LBV with the initial temperature at 400 K. The LBV decrease with the increasing of pressures. Some of the main chain reactions of methanol is very sensitive to pressure, which will slow down the total fuel oxidation rate. With the increase of initial pressure, the chemical reaction rate and LBV of methanol/ethanol/air flames decreases. The correlation formula between LBV and pressure can be described:

$$S_{u0} \propto P^{\frac{n-2}{2}} \quad (5)$$

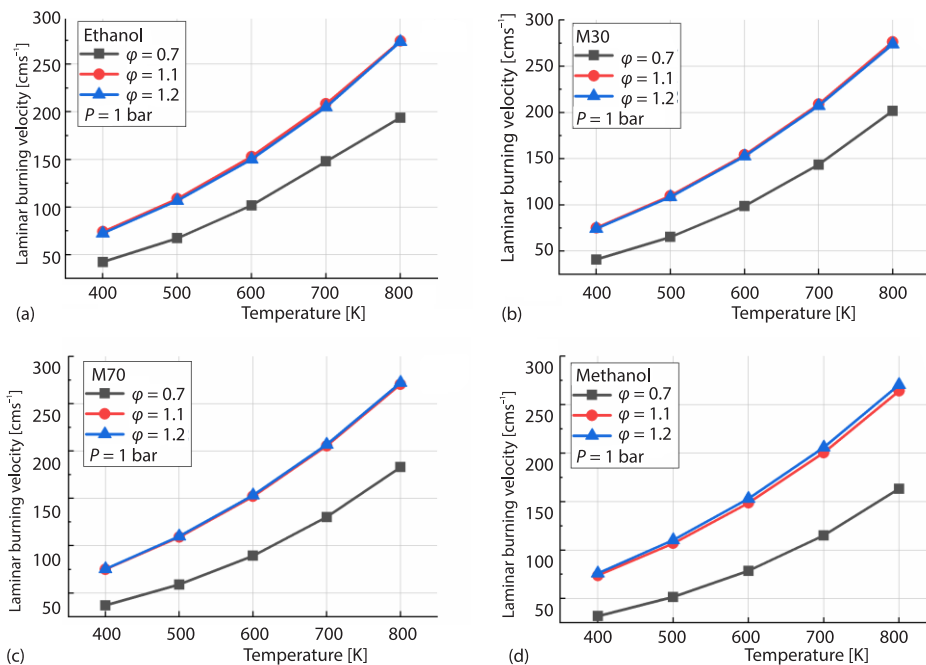
where  $S_{u0}$  is LBV and  $n$  is reaction order ( $n < 2$ ). The numerical value of this equation shows that LBV decreases with the increase in pressure.

Different fuels are affected by the initial pressure differently. Under low pressure, the LBV is greatly affected by pressure. With the increase of methanol content, the fuel has higher pressure sensitivity, which shows that the LBV of methanol decreases more than that of ethanol. Under the condition of high pressure, the LBV changes slowly. Interestingly, only M70's LBV peaks at high pressure when the equivalence ratio is closer to 1.2.

### Flame temperature

Flame temperature reflects the gas mixture's exothermic combustion and the degree of the combustion process [25]. Figure 5 shows the 1-D flame temperature distributions of ethanol, M50, and methanol at different equivalence ratios at an initial temperature of 400 K and an initial pressure of 1 bar. It can be seen from the figure that the flame temperature has reached a higher value at the 0.2 cm position, but is slightly less than the respective adiabatic flame temperature.

Adiabatic flame temperature refers to the heat released by reactant combustion used to raise the temperature of combustion products in an isobaric adiabatic combustion system [26]. However, there will be heat loss, chemical kinetics, or mass transportation limit in real systems, so the flame temperature is often lower than the adiabatic flame temperature [27]. Figure 5(d) shows the adiabatic flame temperature of methanol/ethanol mixed fuel under different equivalence ratios. It can be seen from fig. 5 that the adiabatic flame temperature of the fuel reaches its



**Figure 3. The LBV of different fuels varied with the initial temperature at  $P = 1$  bar; (a) ethanol, (b) M30, (c) M70, and (d) methanol**

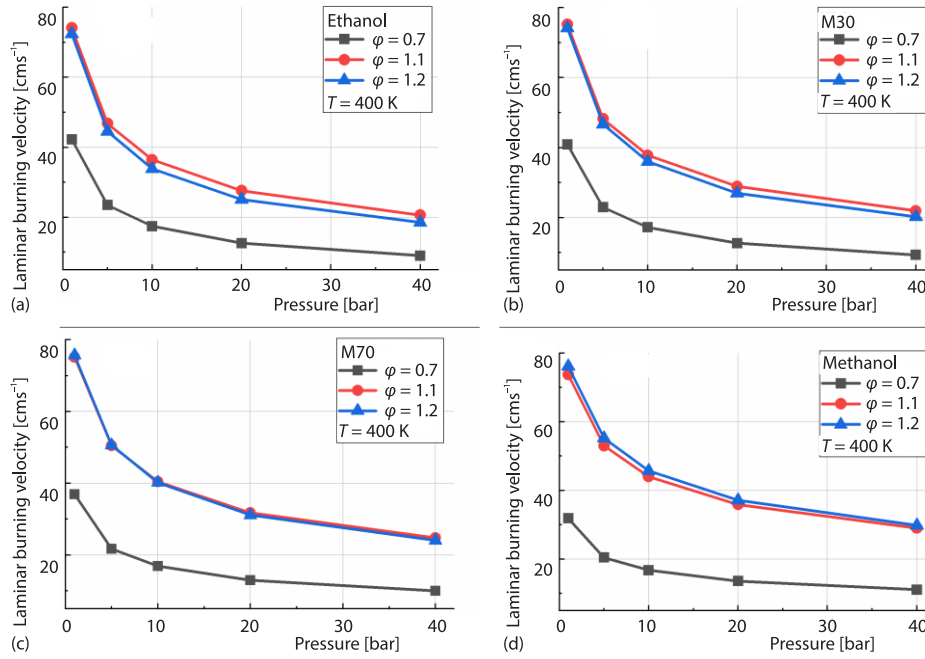


Figure 4. The LBV of (a) ethanol, (b) M30, (c) M70, and (d) methanol at 400 K as a function of pressure

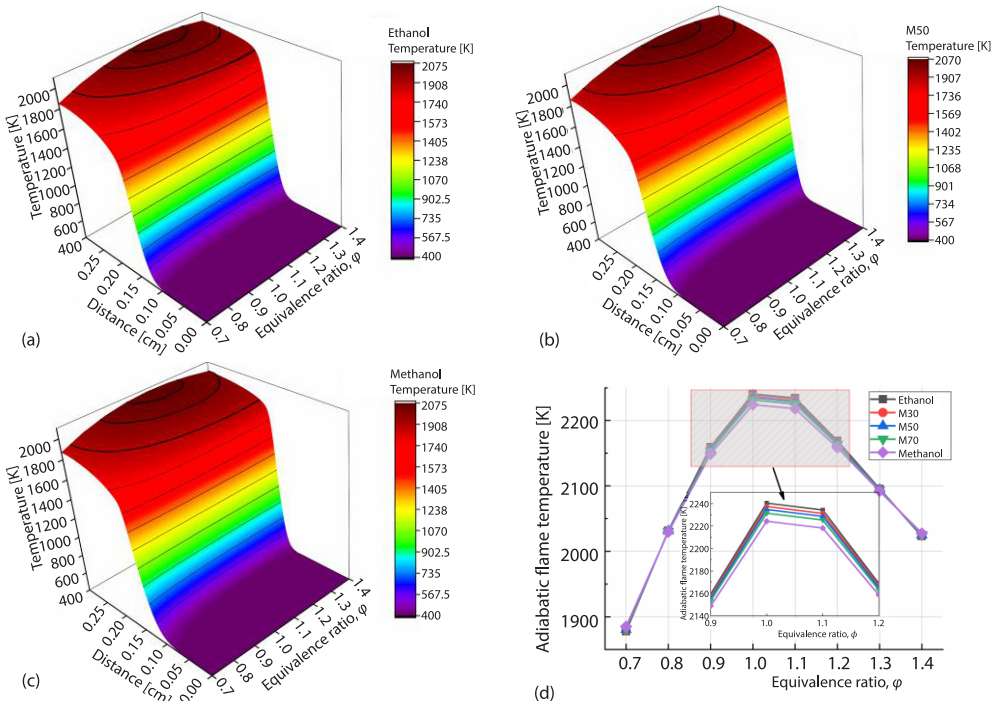


Figure 5. The 1-D flame temperature distribution of methanol/ethanol/air flames; (a) ethanol, (b) M50, (c) methanol, and (d) AFT



peak when the equivalence ratio is 1.0. The adiabatic flame temperature and high temperature flame area of ethanol are higher than that of methanol because of its higher calorific value and oxygen content. When the equivalence ratio is high or low, the difference in the adiabatic flame temperature is smaller (less than the unit equivalence ratio). The slight difference in the adiabatic flame temperature of the mixed fuel proves that there is little difference in spontaneous combustion between methanol and ethanol.

### Heat release

Figure 6 shows the peak net heat release rate (NHRR) and the average heat release rate,  $HRR_0$ , of methanol/ethanol/air flames. The addition of methanol can significantly increase the heat release rate of methanol/ethanol/air flames, and the reaction rate increases significantly also with the increase of methanol content. Moreover, similar to LBV, the position of the peak heat release rate of methanol/ethanol/air flames is closer to the stoichiometric ratio than methanol/air flames.

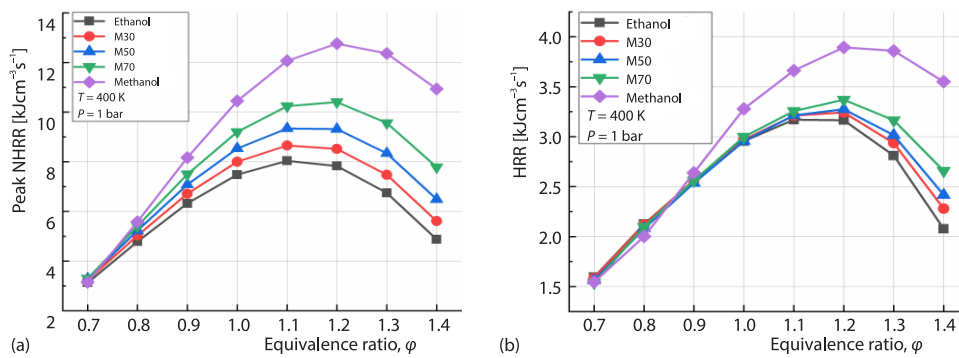


Figure 6. The peak NHRR and  $HRR_0$  of methanol/ethanol/air flames with different methanol content and  $\phi$ ; (a) NHRR and (b)  $HRR_0$

### Flame instability

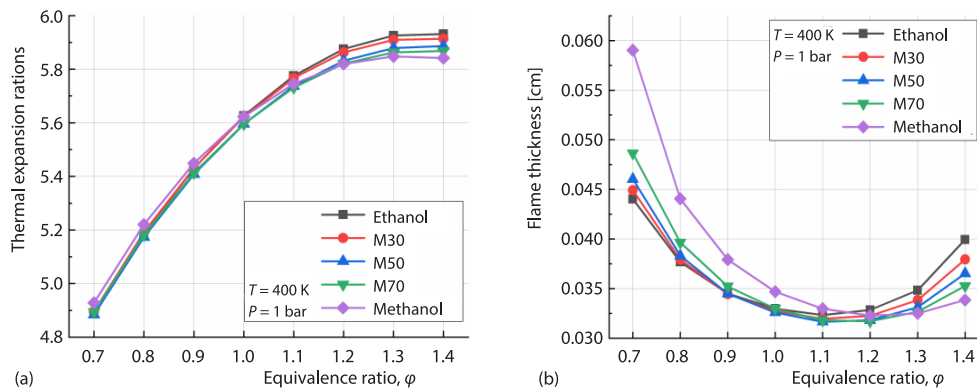
Flame instability is one of the characteristics of laminar premixed flame. Flame instability causes local wrinkles on the flame front, thus affecting the local heat release rate and combustion efficiency. Thermal expansion,  $\sigma$ , and laminar flame thickness,  $\delta$ , are the two main parameters associated with the intensity of hydrodynamic instability. Generally, a higher  $\sigma$  and thinner  $\delta$  correspond to increased hydrodynamic instability intensity [28]. Parameters  $\sigma$  and  $\delta$  are defined:

$$\sigma = \frac{\rho_u}{\rho_b} \tag{6}$$

$$\delta = \frac{T_b - T_u}{\left(\frac{dT}{dx}\right)_{\max}} \tag{7}$$

where  $\rho_u$  and  $\rho_b$  are the unburned and burned gases densities, respectively,  $T_b$  and  $T_u$  – the unburned and burned gases temperatures, respectively, and  $(dT/dx)_{\max}$  is the maximum temperature gradient.

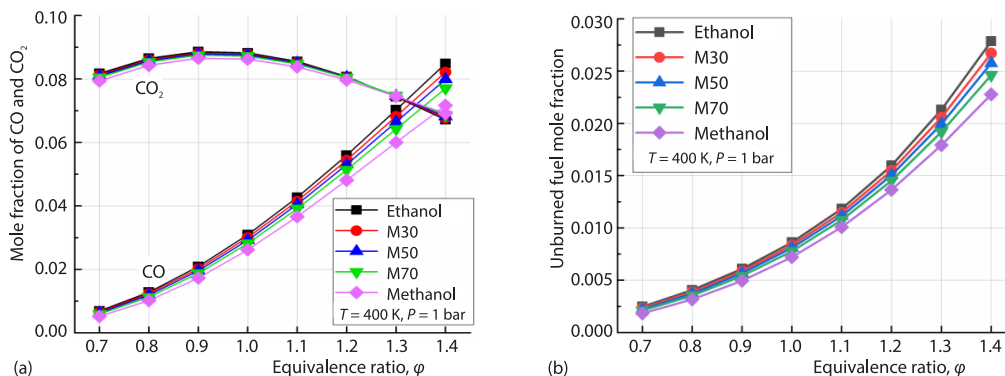
Figure 7 shows the thermal expansion ratios and laminar flames thickness of methanol/ethanol/air flames. The results show that the hydrodynamic instability intensities of ethanol/air flames increase at first and then decrease with the equivalence ratio, reaching the maximum at  $\varphi = 1.1$ . However, with the addition of methanol, the decrease of hydrodynamic instability intensities of methanol/ethanol/air flames are not obvious after  $\varphi = 1.1$ . Both  $\sigma$  and  $\delta$  decrease with the increase of methanol content, so it is impossible to determine whether methanol will increase the hydrodynamic instability intensities of methanol/ethanol/air flames. It is considered that the flame stability of methanol is similar to that of ethanol.



**Figure 7. The thermal expansion ratios and laminar flames thickness of methanol/ethanol/air flames; (a) thermal expansion ratios and (b) laminar flames thickness**

### The CO, CO<sub>2</sub>, and unburned fuel

Generally speaking, the exhaust gas from the combustion of alcohol fuels inevitably contains CO, CO<sub>2</sub>, and unburned fuels, which are harmful to the environment. Therefore, it is necessary to reduce these pollutant emissions. The mole fraction of CO and CO<sub>2</sub> at the end of the calculation domain for various methanol components and  $\varphi$  is shown in fig. 8(a). It was found that adding methanol to ethanol can decrease CO and CO<sub>2</sub> emissions. In addition, CO emission increases rapidly with the increase of  $\varphi$ . The CO<sub>2</sub> emissions increase first and then decrease with the increase of  $\varphi$ , but the range of change is significantly smaller than that of CO emissions.



**Figure 8. The mole fraction of CO, CO<sub>2</sub>, and unburned fuel at the endpoint of methanol/ethanol/air flames; (a) mole fraction of CO and CO<sub>2</sub> and (b) unburned fuel mole fraction**



Figure 8(b) shows the unburned fuel mole fraction in the exhaust gas with different  $\phi$  and methanol components. It can be seen that the mole fraction of the unburned fuel increases greatly with increasing the equivalence ratio. Adding methanol reduces the emission of the unburned fuel. Therefore, considering the emission of CO, CO<sub>2</sub>, and unburned fuel comprehensively,  $\phi < 1$  is suitable for the combustion of methanol/ethanol flame.

### Flow rate sensitivity

Sensitivity analysis is to find the key reactions in the combustion process and analyze their effects on the characteristics of laminar flames. If a certain reaction has a positive coefficient, it means that the reaction has an increasing influence on the characteristic. On the contrary, a negative coefficient means that the influence of the reaction on the characteristic decreases [29].

Figure 9 shows the main reactions of M50 affecting the LBV at different temperatures. The reaction involved is the combination reaction between hydrocarbon mixtures, which produces or consumes O, H, O<sub>2</sub>, H<sub>2</sub>, OH, and H<sub>2</sub>O. The results show that the system produces H or OH radicals, and H and OH are the two main elements that increase the LBV. The reaction R1 has the greatest promotion effect on LBV. The reaction R27 is also a powerful carrier of the LBV because CO is also a good fuel. However, CO<sub>2</sub> is a product that inhibits the combustion process. The R12 and R30 also have positive effects on the LBV.

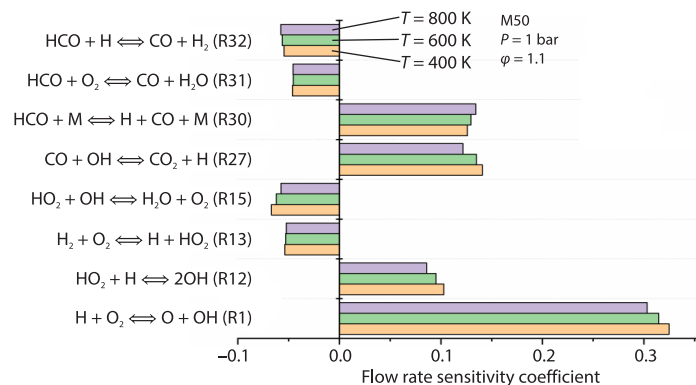


Figure 9. Main influential reactions of different temperatures at  $\phi = 1$  (M50)

### Conclusions

The laminar burning velocity of different fuels under different conditions was simulated by Chemkin software. The laminar burning velocity, 1-D flame temperature, flame instability, pollutant emission, and RoP analysis were studied. The main conclusions are as follows.

- With the increase of methanol component, the equivalence ratio at peak LBV of the blended fuel increases from 1.1-1.2. At the same time, the LBV of the blended fuel is proportional to temperature and inversely proportional to pressure. The adiabatic flame temperature of the fuel reaches its peak at the equivalence ratio of 1.0. The adiabatic flame temperature and high temperature flame area of ethanol are higher than that of methanol because of their higher calorific value and oxygen content. The addition of methanol can significantly increase the heat release rate of methanol/ethanol/air flames, and the reaction rate increases significantly with the increase of methanol content. In addition, similar to the LBV, the position of the peak heat release rate of methanol/ethanol/air flames changes with the addition of methanol.

- The hydrodynamic instability intensity of ethanol/air flames first increase and then decrease with the equivalence ratio, and reaches the maximum at  $\varphi = 1.1$ . Both  $\sigma$  and  $\delta$  decrease with the increase in methanol, and it is considered that the flame stability of methanol is similar to that of ethanol. Adding methanol can reduce the emission of CO, CO<sub>2</sub>, and unburned fuel. At the same time, excessive air combustion (while  $\varphi < 1$ ) significantly reduces pollutant emissions. Flow rate sensitivity varies with temperature.  $\text{H} + \text{O}_2 \rightleftharpoons \text{O} + \text{OH}$  (R1) reaction is the key reaction affecting the LBV.

This research indicates that methanol and ethanol have similar laminar combustion characteristics, at the same time methanol can improve the combustion performance of ethanol–air mixture and reduce pollutant emissions. The research results enrich the basic data of methanol/ethanol/air flame in the process of laminar premixed combustion and provide a basis for the design of methanol-mixed ethanol engines.

### Acknowledgment

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### Nomenclature

$AFT$  – adiabatic flame temperature, [K]  
 $LBV$  – laminar burning velocities, [ $\text{ms}^{-1}$ ]  
 $\dot{m}$  – mass-flow, [ $\text{kg s}^{-1}$ ]  
 $P$  – pressure [bar]  
 $S_{U,0}$  – laminar burning velocity, [ $\text{ms}^{-1}$ ]  
 $T$  – temperature, [K]  
 $T_{u,0}$  – reference temperature, [K]  
 $\bar{W}$  – mean molecular weight, [ $\text{gmole}^{-1}$ ]

### Greek symbols

$\delta$  – laminar flame thickness, [mm]  
 $\lambda$  – thermal conductivity, [ $\text{W m}^{-1} \text{K}^{-1}$ ]  
 $\rho$  – density of mass, [ $\text{kg m}^{-3}$ ]  
 $\sigma$  – thermal expansion, [–]  
 $\varphi$  – equivalence ratio, [–]

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