# THE TEMPERATURE DEPENDENCE OF LAMINAR BURNING VELOCITIES OF METHANOL-SYNGAS-AIR FLAMES

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ABSTRACT

Using the simplest alcohol – methanol – as a fuel for spark ignition (SI) engines, enables an increase of thermal efficiency compared to gasoline. Additionally, with the enrichment of hydrogen rich gas from methanol reforming (syngas) using exhaust heat, the efficiency can be further improved. The complexity of optimizing such an arrangement asks for numerical support. However, there is no research that publishes the effect of unburned mixture temperature and equivalence ratio on the laminar burning velocity of methanol-syngas blends, which is needed for developing an engine cycle code to simulate methanol fueled SI engines with syngas addition from exhaust gas fuel reforming.
The influence of temperature on the laminar burning velocity of methanol-syngas blends is investigated in this study using CHEM1D. The simulation shows that the flame speed increases dramatically with the enrichment of syngas, especially at lean and rich conditions. The effect of syngas ratio on the improvement of burning velocity is less important at higher temperatures, and there is almost no influence at stoichiometry.
Some well-known mixing rules are then examined. In general, the Hirasawa mixing rule shows the best fit with the numerical data. For blends with high syngas content, the Le Chatelier's mixing rule is recommended. The temperature power exponent α is calculated and compared to other correlations. It shows that the published correlations are unable to predict the influence of temperature on laminar burning velocity accurately enough for the combustion of methanol, syngas and their blends in air.

TECHNICAL PAPER

INTRODUCTION

Internal combustion engines (ICEs) play an important role in the development of modern society, especially in the transportation sector. In the near future, the ICE will still be the main type of powertrain for vehicles, but the energy carrier it uses should increasingly come from renewable fuels, like methanol ($CH_3OH$) [1]. Several researches show the potential of methanol as a fuel for ICEs to improve the thermal efficiency and reduce pollutant emissions [2-4]. The improvement is observed due to a variety of interesting properties of methanol compared to gasoline, as shown in Table 1. With high heat of vaporization and low air to fuel ratio, the intake charge is cooling down when methanol evaporates, which leads to increasing the engine volumetric efficiency. Additionally, with the cooler intake charge, higher octane number and faster flame speed, using methanol as a fuel reduces the possibility of abnormal combustion, knock, to occur in SI engines. A higher compression ratio, and less heat losses, further improves the engine thermal efficiency. Methanol is also a liquid fuel, easy for storage, distribution and possibly for the evolution of current infrastructure.

| Property | Gasoline | Methanol | Hydrogen | Carbon monoxide |
|---|---|---|---|---|
| Chemical formula | Various | $CH_3OH$ | $H_2$ | $CO$ |
| Lower heating value [MJ/kg] | 42.9 | 20.1 | 120 | 10.1 |
| Stoichiometric air to fuel ratio [kg/kg] | 14.7 | 6.5 | 34.32 | 2.45 |
| Research octane number (RON) [–] | 92 – 98 | 109 | 130 | 106 |
| Boiling point at 1 bar [°C] | 25 – 215 | 65 | -253 | -191.5 |
| Heat of vaporization, $\phi = 1$ [kJ/kg Air] | 18 | 169.2 | – | – |
| Flammability limit in air [vol.%] | 1.2 – 7.1 | 6.7 – 36 | 4 – 75 | 12.5 – 74 |
| Minimum ignition energy in air [mJ] | 0.8 | 0.14 | 0.017 | < 0.3 |
| Laminar flame speed, $\phi = 1$ [cm/s] | 28 | 42 | 210 | 14.5 |

Table 1: Properties of gasoline, methanol, hydrogen and carbon monoxide at NTP [3-6]

Thanks to its high H/C ratio, methanol is also considered as a hydrogen carrier fuel. Due to the strict requirements of storage and distribution of compressed gas/liquid hydrogen, methanol reforming technology has been proposed to produce hydrogen rich gas (syngas) onboard the vehicle. The main application of syngas now is in industries, especially as a fuel for gas turbines. The literature contains many investigations into the laminar burning velocities of syngas with different $H_2/CO$ ratios [7-10]. The laminar flame speed increases significantly with higher hydrogen ratio, and much faster than that of conventional liquid fuels for SI engines. Therefore, some studies confirmed that the engine performance was enhanced with the addition of hydrogen or hydrogen rich gas into the intake manifold [11-13]. As the required temperatures for the methanol reforming process are much lower than that of other fuels like ethanol and gasoline, some studies have used methanol as a fuel for the on-board reformer together with gasoline for their engine [14, 15]. Leslie Bromberg *et al.* [16] concluded that the efficiency of methanol fueled SI engine can be further improved, up to 50-60%, with the addition of syngas from the on-board reformer driven by engine exhaust heat. The improvement of engine efficiency is due to the high flame speed, high octane number of syngas and its flammability limits. The engine is able to operate under higher compression ratio, extremely lean condition, or high dilution ratio. However, there has been no research publishing the laminar burning velocity of methanol-syngas blends in air, which is needed to develop the correlation for engine cycle codes to reduce the time and money for engine optimization. This paper investigates the influence of temperature on the laminar burning velocity of methanol-syngas blends in air using the correlation $u_L = u_{L0}.(T_u/T_0)^\alpha$ with the help of a one-dimensional flame code. Some published mixing rules are also examined in this study.

NUMERICAL METHODOLOGY

The comprehensive reaction mechanism developed by Li *et al.* [17] was used in this study. Although it was updated from the methanol oxidation mechanism [18], to be able to simulate the combustion of $CO/H_2/H_2O/O_2$ and $CH_2O$, Li's mechanism was one of the five best mechanisms for syngas oxidation [19]. Therefore, it was selected to predict the laminar burning velocities of methanol, syngas and their blends in air. Five fuels (MeOH, syngas, MeOH25, MeOH50 and MeOH75) were tested in this study. The "MeOH" designates methanol and the number next to "MeOH" designates the percentage by volume of methanol in the fuel blends. Assuming the fuel conversion is 100%, there are three main products of methanol steam-reforming, after condensation (CO, $CO_2$ and $H_2$). However, their concentration varies for different conditions. Their ratio is a function of the CO selectivity (from 0% to 100%). Additionally, because of the lack of experimental data for $H_2/CO/CO_2$ blends for the validation, in this research, syngas was simplified to a mixture of only two components, hydrogen and carbon monoxide. The selected syngas has a ratio by volume of

H$_2$/CO being 50/50. This syngas has a lower heating value (LHV) similar to that of an H$_2$/CO/CO$_2$ mixture with CO selectivity of 50%. The volume fraction of the three components of the five tested fuels and their LHVs are plotted in Fig. 1.
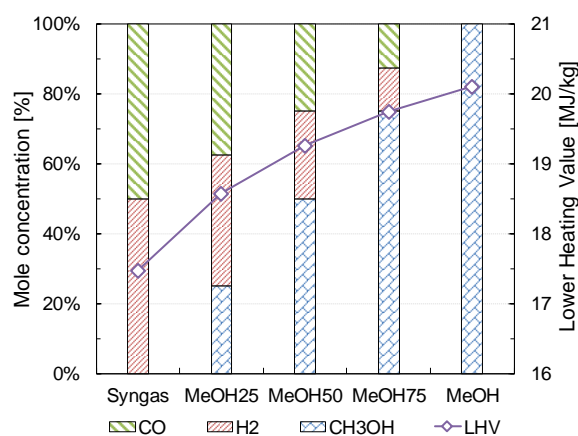


Figure 1: The component of tested fuels, their concentrations by volume and lower heating values

In order to determine the laminar burning velocity of the tested fuels, a one-dimensional flame code – CHEM1D was used. This code was developed by the Combustion Technology group, at Eindhoven University of Technology. In each case, the stationary simulation was performed with an exponential differential scheme and free flame type using 200 grid points. The studied range of equivalence ratio was 0.5 – 2.0 with $\Delta\phi$ = 0.1, and unburned gas temperature was 300 K – 900 K ($\Delta T$ = 50 K) at atmospheric pressure. In total, 1040 independent simulations were performed.

MIXING RULES

For the fuel blends, it is essential to know how the laminar burning velocity varies when the blend ratios are changed. However, most of the current reaction mechanism was developed and validated for a single fuel. Developing a comprehensive mechanism for a mixture of several fuels requires a lot of works and computation time. Knowledge of that behavior without chemical kinetic calculations could allow faster simulation of the engine. In order to find a mixing rule to predict the laminar burning velocity of methanol-syngas blends, an examination of existing mixing rules was done. Five mixing rules are evaluated in this study, including mole fraction, mass fraction, energy fraction [20], Le Chatelier's [21] and Hirasawa's mixing rules [22]. According to Sileghem *et al.* [20], the energy fraction mixing rule, the mixing rule developed by Hirasawa *et al.*, and the Le Chatelier's rule performed very well for blends of hydrocarbons and ethanol. However, these mixing rules did not work for hydrogen-methane blends because of the strong reactivity of hydrogen. In the discussion below, these mixing rules have been appraised for the blends of methanol and syngas.

RESULTS AND DISCUSSION

Figure 2 shows the validation of laminar burning velocities of methanol-air and syngas-air at atmospheric pressure and initial temperature of around 300 K. The comparison showed that the calculated $u_L$ with Li's mechanism fits well with the experimental data from literature for both methanol-air and syngas-air flames. Although Sun *et al* [9] concluded that Li's mechanism over predicts for syngas-air mixtures at $\phi > 2.0$, with the range of $\phi$ in this study, the results from CHEM1D generated results with a fairly high level of accuracy.
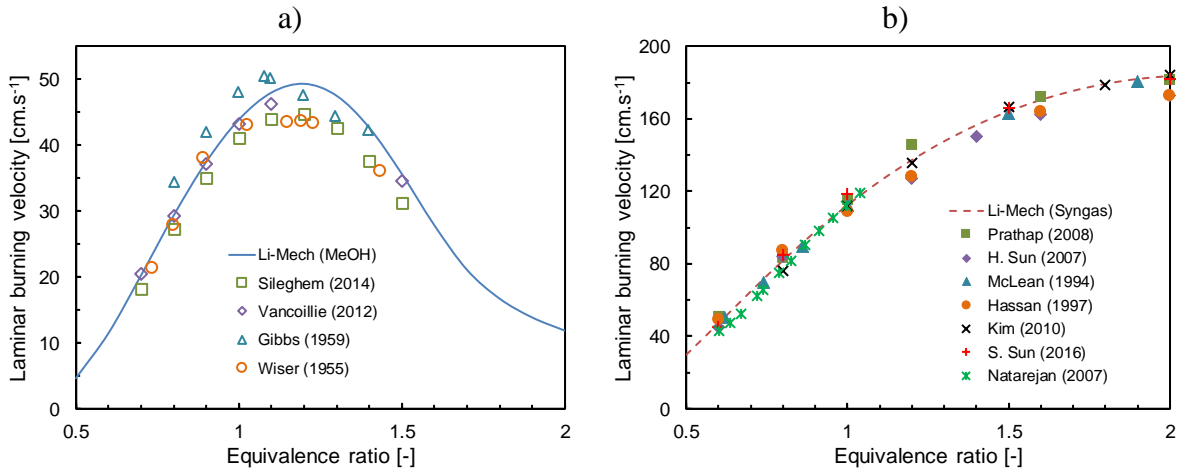
Figure 2: Validation of $u_L$ of methanol (a) and syngas (b) at $p$ = 1 bar and $T_u$ of ~300 K. Lines – CHEM1D results with Li's mechanism, opened symbols – experimental data of methanol-air flames [23-26], closed symbols – experimental data of syngas-air flames [9, 10, 27-31]
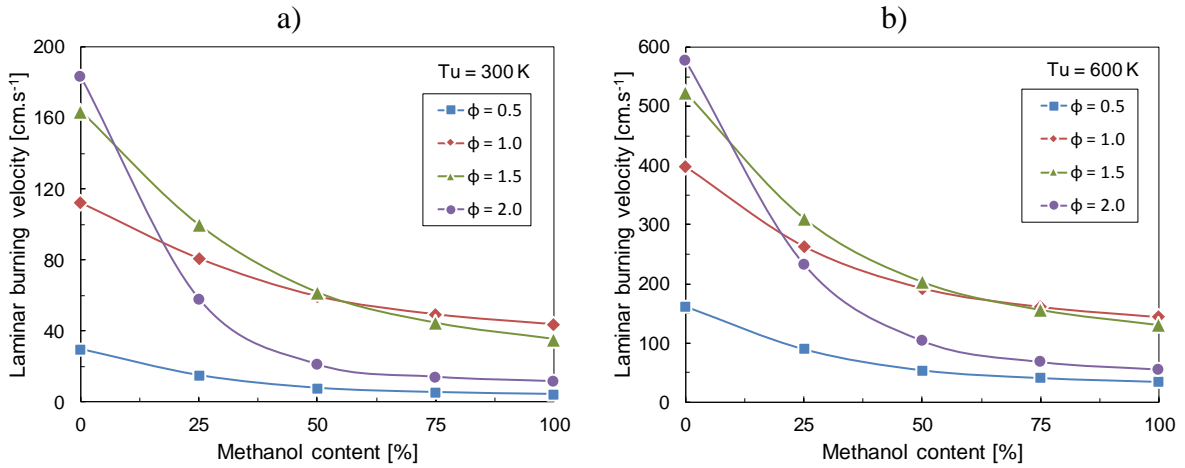


Figure 3: Influence of syngas contents in the mixtures on $u_L$ at $p$ = 1 bar, $T_u$ of 300 K (a) and 600 K (b)
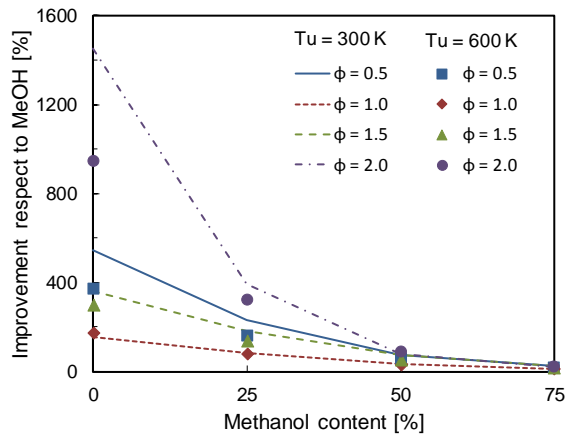


Figure 4: The improvement of laminar burning velocity of methanol-syngas mixtures versus methanol contents (respected to pure methanol) at $p$ = 1 bar. Lines – $T_u$ of 300 K, symbols – $T_u$ of 600 K

Figure 3 presents the laminar burning velocities of all tested fuels in air as a function of methanol contents in the mixture at four different equivalence ratios, and $T_u$ of 300 K and 600 K. The laminar burning velocity of methanol-syngas blends significantly increases with the reduction of methanol content (or the increase of syngas ratio). Figure 4 displays the improvement of laminar burning velocity with respect to pure methanol as a function of

methanol content at four equivalence ratios and two unburned temperatures. It can be seen that the syngas addition and unburned temperatures have fewer influence at stoichiometric condition. It explains why the effect of hydrogen addition on engine performance is most obvious at lean conditions [32].

The data of MeOH50-air flames then were compared to the calculated values using different mixing rules, as shown in Fig. 5. It is clear that mole, mass and energy fraction mixing rules were unable to accurately predict the laminar flame speed of the methanol-syngas blend accurately, especially at rich conditions. This is due to the fact that the peak burning velocities of methanol and syngas occur at different $\phi$. The fastest laminar burning velocity of methanol-air was observed at $\phi = 1.2$, whereas the equivalence ratio for the peak burning velocity of syngas-air is 2.0. This difference results from the high thermal and mass diffusivities of hydrogen compared to normal liquid fuels.
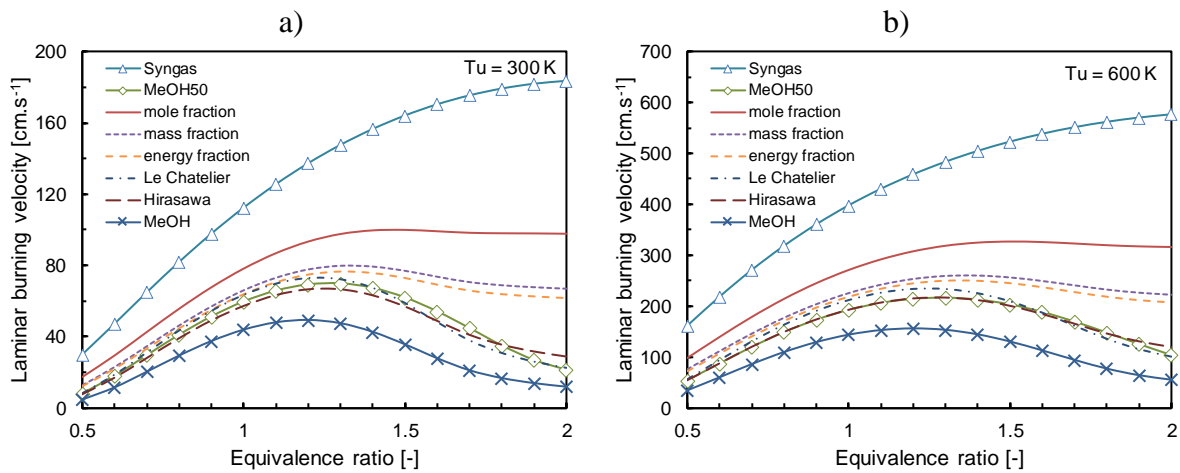


Figure 5: Simulated $u_L$ and calculated $u_L$ as a function of $\phi$ using different mixing rules of MeOH50-air flames at $p = 1$ bar, $T_u$ of 300 K (a) and 600 K (b)
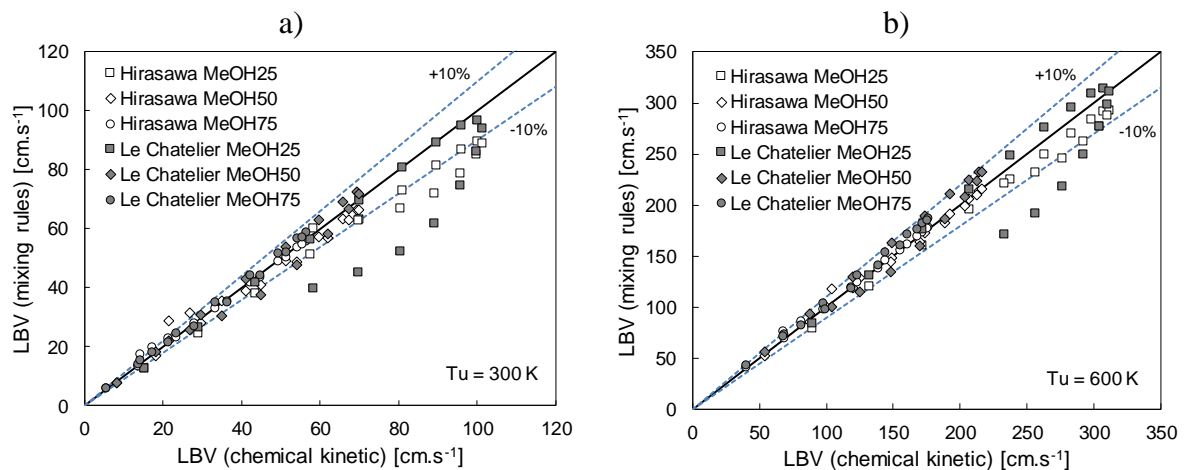


Figure 6: Laminar burning velocity of methanol-syngas blends: mixing rules versus kinetic calculation at all equivalence ratios and two temperatures, $T_u = 300$ K (a) and $T_u = 600$ K (b)

| Fuel | $T_u = 300$ K | | $T_u = 600$ K | |
| --- | --- | --- | --- | --- |
| | Le Chatelier | Hirasawa | Le Chatelier | Hirasawa |
| MeOH25 | 14.19 (1.72) | 10.18 (5.91) | 30.5 (8.18) | 18.8 (11.85) |
| MeOH50 | 3.35 (2.09) | 3.59 (1.66) | 12.52 (12.98) | 4.47 (0.66) |
| MeOH75 | 1.39 (1.41) | 1.24 (0.37) | 6.51 (7.04) | 2.6 (0.65) |

Table 2: RMSE of the different mixing rules. The data in bracket presents RMSE for $\phi \leq 1$ (cm.s$^{-1}$)

Le Chatelier's and Hirasawa's mixing rules predict the burning velocity of methanol-syngas blends well. In general, the Hirasawa mixing rule provides better results than the Le Chatelier's rule. Especially, at unburned gas temperature of 600 K, the data from the calculation with Hirasawa mixing rule shows perfect fit with the simulated results. Figure 6 indicates the comparison of laminar burning velocities which were calculated using Le Chatelier and Hirasawa mixing rules versus the kinetic simulation results at two temperatures, 300 K and 600 K. It is clearly seen that Le Chatelier and Hirasawa mixing rules are only predictive for the mixture with syngas ratios less than or equal to 50%. If the allowable deviation is 10%, the Hirasawa mixing rule is preferred to predict the flame speed of methanol-syngas blends at high temperature, and for lean and stoichiometric mixtures at low temperature. However, Le Chatelier's mixing rule is recommended for the prediction the flame speed of the mixture which has high syngas content (75%) at lean and stoichiometric conditions.
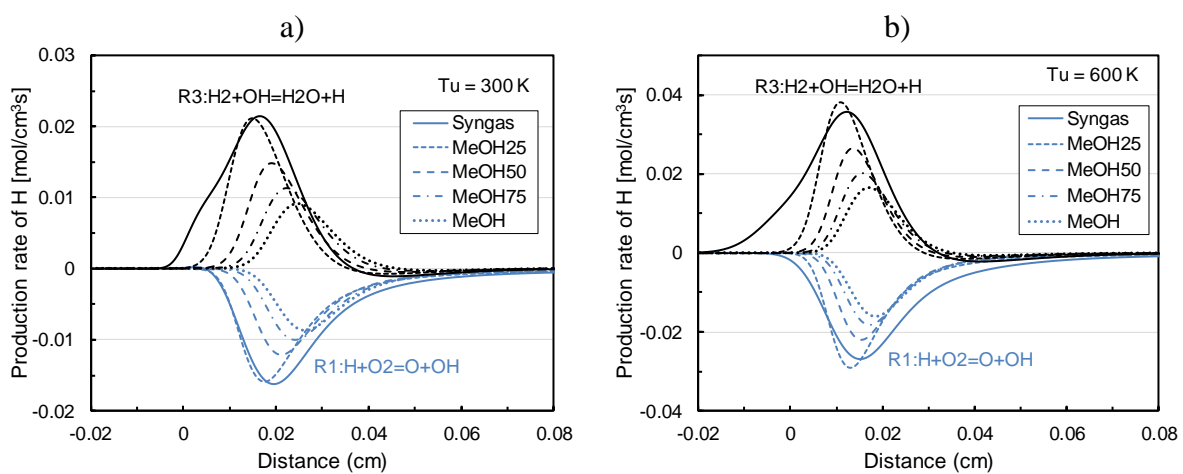


Figure 7: Production rate of H for different blends at $\phi = 1$, $T_u = 300$ K (a) and $T_u = 600$ K (b)
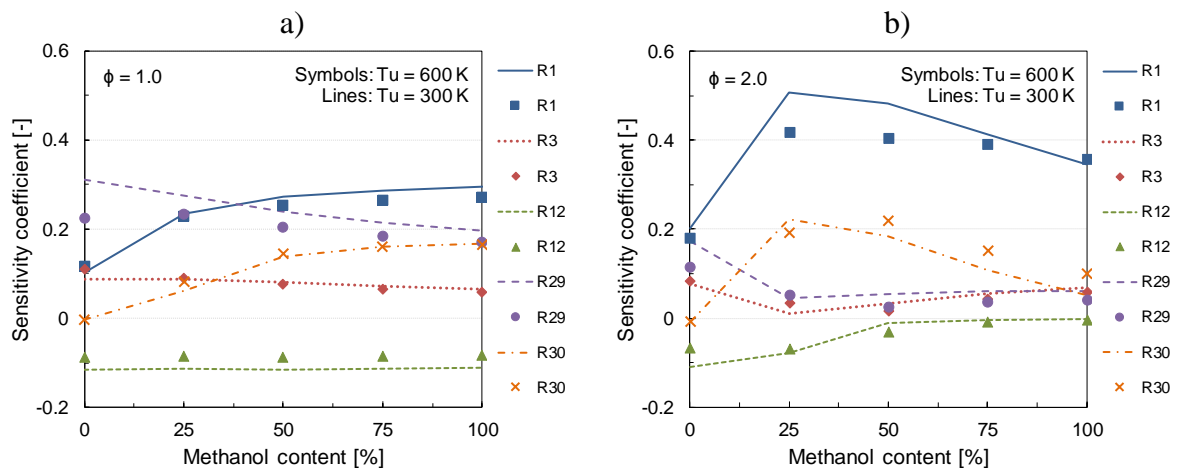


Figure 8: Sensitivity coefficients of dominant reactions of mass burning rate versus methanol blend ratios at two equivalence ratios, $\phi = 1.0$ (a) and $\phi = 2.0$ (b)

Table 2 displays the root-mean-square-error (RMSE) of the two mixing rules compared to the data from the chemical kinetic calculations. The data in the brackets is the RMSE for lean and stoichiometric mixtures. There is a trend that the RMSE becomes larger with less methanol (more syngas) content for both mixing rules, which is due to the kinetic interactions having a larger influence on the flame speed. As shown in Fig. 7, the peak production rate of H

increases dramatically with more syngas in the blends, and there is no linear relationship between the methanol/syngas ratio and the peak production rate of the H radical.

Figure 8 presents the sensitivity coefficients of five dominant reactions of mass burning rate versus methanol blend ratios at two equivalence ratios, $\phi = 1.0$ and $\phi = 2.0$. In general, two reactions R1 ($H+O_2=O+OH$) and R29 ($CO+OH=CO_2+H$) are the most dominant reactions which influence the mass burning rate at stoichiometric condition. For lean mixtures, R29 becomes a less important reaction. At $\phi = 1.0$, there was almost no influence of methanol content on the sensitivity coefficients for the mixtures of which the methanol content was greater than 50%. This is the reason why the influence of methanol ratio on the improvement of flame speed (as shown in Fig. 4) becomes less important at $\phi = 1.0$.

Figure 9 shows the calculated α based on the correlation $u_L = u_{L0}.(T_u/T_0)^\alpha$. It is clearly seen that for a certain equivalence ratio, a single value of α is not able to present the influence of unburnt temperature on flame speed. The α derived from the slope of line fitting in the log-log plot is equal to the calculated α at maximum temperature. The laminar burning velocity at lower temperatures calculated with that α is higher than the observed value. For this reason, the averaged α was used to represent the temperature power exponent.
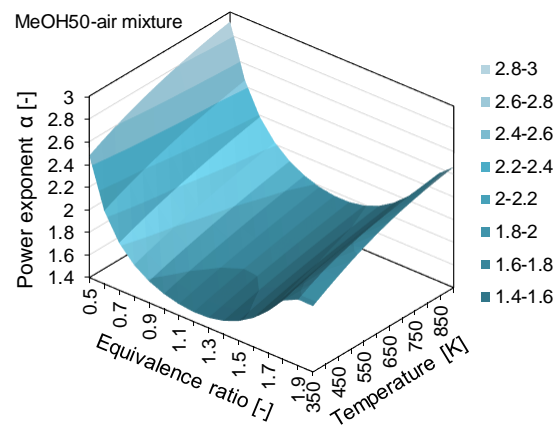


Figure 9: The map of power exponent α of MeOH50-air mixture as a function of $\phi$ and $T_u$ at $p = 1$ bar
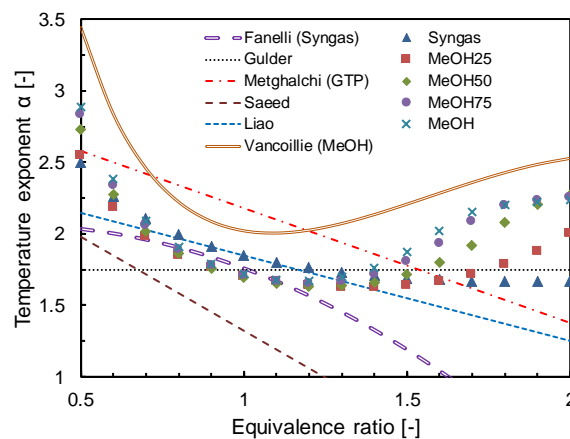


Figure 10: Comparison between temperature exponent α from simulation and α from published correlations

The averaged α values of all fuels are plotted together with data from other research in Fig. 10. There is no significant difference between the α of methanol and methanol-syngas blends at close to stoichiometric conditions. It is also clearly seen that the well-known correlations for methanol-air flames like Metghalchi & Keck [33], Liao [34] and Saeed [35] are not able to

predict the laminar burning velocities at high temperatures for all tested fuels. The Gulder model [36] is only predictive for mixtures at stoichiometric condition. The α from the correlation developed by Vancoillie *et al.* [37] is over predicting. Fanelli *et al.* [38] developed a correlation for the laminar flame speed of syngas, but their α is only acceptable for the mixtures which are close to the stoichiometric condition. Therefore, there is a need for the development of a new laminar burning velocity correlation for methanol and methanol-syngas blends to implement it into the engine simulation tool.

CONCLUSIONS

This paper showed the influence of unburned temperatures on the flame speed of methanol-syngas blends in air at atmospheric pressure using CHEM1D. The calculations were done in a wide range of equivalence ratios (0.5 – 2.0), and temperatures (300 K – 900 K).

The simulation showed that the laminar burning velocity of fuel blends increased dramatically with higher syngas contents. The influence of the syngas ratio became less important at stoichiometric conditions and at higher temperature. Several mixing rules were examined for the prediction of laminar burning velocities of methanol-syngas blends in air. The mass fraction, mole fraction and energy fraction rules showed a worse prediction compared to Le Chatelier and Hirasawa rules. In general, the Hirasawa mixing rule offered a better prediction compared to that of Le Chatelier. However, Le Chatelier's mixing rule was recommended for the mixture with high content of syngas.

The averaged power exponents α at each equivalence ratio were calculated and compared with other α from developed correlations. Only a few correlations are predictive for the mixtures close to the stoichiometric condition. A new laminar burning velocity correlation of methanol and methanol-syngas blends is required for use in engine cycle codes. Further validation of the correlation also needs to be done, e.g. through experimental studies using spherically expanding flames in a constant volume combustion chamber.

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