

## Temperature Dependence of Laminar Burning Velocity in Ammonia/Dimethyl Ether-air Premixed Flames

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**Abstract:** The combustion of ammonia ( $\text{NH}_3$ ) has attracted wide interest in fuel vehicle engines, marine engines, and power generators to mitigate carbon dioxide emissions. Unfortunately, the relatively low laminar flame speed presents a technical barrier for this renewable fuel to be used in practice. This work is concerned with numerical examining the effects of elevating inlet temperature on the laminar burning velocity of  $\text{NH}_3$ /air flames with various contents of dimethyl ether (DME) using 1D freely propagating flame calculations, and to shed light on the flame enhancement mechanism. For this, the mechanism is first validated by comparing the numerical predictions with experimental data. Results show that increasing the inlet temperature has a positive effect on the laminar burning velocity of pure  $\text{NH}_3$ /DME/air flames. It is revealed that elevating inlet temperature contributes to a higher adiabatic flame temperature, which is beneficial to the overall chemical reaction rate. Furthermore, the thermal diffusivity of the binary mixture is observed to increase substantially as well. Further kinetic and sensitivities analyses reveal that the inlet temperature has a minimal effect on the reaction pathway, leading to the relative importance of the dominant chain branching over terminating reaction steps to be varied negligibly. The present work confirms that the flame speed enhancement with increasing inlet temperature is primarily the synergetic result of the thermal and diffusion effects, rather than the chemical effect.

**Keywords:** ammonia, dimethyl ether, laminar burning velocity, temperature dependence, kinetic analyses

### 1. Introduction

As climate change has become increasingly severe and started affecting human being's life, there has been a surge of interest and demand in seeking alternative fuels for power and propulsion systems to mitigate carbon dioxides emissions [1–3]. Ammonia, as a renewable and sustainable fuel, is now being recognized as a potential candidate to replace conventional fossil fuels [4, 5]. This interest is stimulated by the following merits: first, ammonia has a high hydrogen content, and its

combustion products are nitrogen and water when complete combustion occurs; second, there are well-established infrastructures for ammonia storage and transport, suggesting that this fuel can be readily accessed without having a high cost; last but not least, some attempts using renewable energy such as wind and solar energy to produce ammonia is underway [6], and some progress has been achieved. Despite these advantages of burning ammonia, its relatively low flame speed has been a great challenge for the wide realization in practical applications. Consequently, it is essential and

significant to address this issue, and this has been a research focus among ammonia-based communities [7–9].

To tackle the poor combustion characteristics of ammonia-involved combustion, some viable strategies have been proposed and tested. One way to achieve this goal is based on varying the inlet thermodynamics [10, 11]. Experiments adopting a constant-volume cylindrical combustion chamber confirmed that increasing the oxygen content can give rise to a considerable increase in the laminar burning velocity [12]. It was demonstrated that the thermal effect which was advantageous for generating more radicals (such as OH and H) played a dominant role in the enhanced flame speed, instead of the chemical effect. Li et al. [13] pointed out that the laminar burning velocity of ammonia/air flames can be increased by up to 2.6 times when the oxygen content was varied from 21% to 30%. Both experimental measurements and numerical predictions indicate that increasing the oxygen content contributes to a high laminar burning velocity of ammonia flames primarily as a consequence of the higher adiabatic flame temperature.

Furthermore, the initial temperature is found to play a vital role in the laminar burning velocity for chemically reacting mixtures. The temperature dependence of the laminar burning velocity for NH<sub>3</sub>/air flames was experimentally and numerically determined [14]. Duynslaegher et al. [15] demonstrated that increasing the initial temperature can contribute to a high flame propagation speed. Experiments assessing the impact of the initial temperature on the flammability limit of ammonia/air mixtures were conducted in a cylindrical combustion vessel [16]. It was found that increasing the inlet temperature can enable the flame flammable range to be extended. Similar results were also reported by Kondo et al. [17]. In addition, nonequilibrium plasma-assisted combustion has been proven to be effective as a flame enabler. Shioyoke et al. [18] pointed out that both the preheating temperature and nonequilibrium plasma-assisted combustion played a positive role in enhancing the flame speed, but the former one tended to exert a stronger effect as a consequence of the unburned gas expansion.

Another commercially used method is based on the concept of blended combustion [19–21]. This method does not involve modifying the combustion chamber and is typically achieved by adding some fuels with relatively high mass diffusivities like hydrogen and methane [22, 23]. Li et al. [24] numerically reported that the existence of hydrogen promotes the dominant two-order reactions, leading to low activation energy and large transport diffusion, thus an increased flame speed. Xiao et al. [25] found that adding ammonia played a negative role in the laminar burning velocity of methane flames. More

recently, it is confirmed that the laminar flame speed of ammonia can be also increased via blending with oxygenated alternative fuel alcohols such as methanol and ethanol, and ethers [26, 27].

The prior studies have noted the importance of enhancing ammonia combustion by increasing the inlet temperature and blending other combustible fuels. However, to the best knowledge of the present authors, there are no experimental or numerical studies assessing the effect of inlet temperature on the NH<sub>3</sub>/dimethyl ether (DME)/air premixed flames. This partially motivates the present study. The objectives of this work are to evaluate this implementation effect, and to identify the mechanism in terms of thermal, diffusion, and chemical effects. Section 2 presents the kinetic modeling process. The effects of inlet temperature and blended ratio on the laminar flame speed are examined, and a detailed discussion is undertaken in terms of reaction path and sensitivity analyses in Section 3. The main conclusions obtained from this work are summarized in Section 4.

## 2. Detailed Kinetic Modeling

### 2.1 Modeling of the premixed laminar burning velocity

This work considers the detailed chemical kinetic mechanism proposed by the Combustion Research Group at the University of California at San Diego to describe the NH<sub>3</sub>/DME oxidation process [28]. This mechanism contains 73 species and 325 reaction steps. The laminar burning velocity is one of the fundamental properties in characterizing a combustible mixture for a given temperature, pressure, and fuel composition for premixed flames [29]. The calculation of such property is performed using 1D freely propagating flame sub-model in ANSYS Chemkin-Pro 2019. The effects of the blended ratio and inlet temperature on the flame speed are evaluated. Hereafter, the blended ratio is defined as the ratio of DME mole fraction over the total fuel mole fraction, which can be written as

$$\xi = \frac{X_{\text{DME}}}{X_{\text{NH}_3} + X_{\text{DME}}} \quad (1)$$

where  $X_{\text{DME}}$  and  $X_{\text{NH}_3}$  represent the mole fractions of DME and NH<sub>3</sub> in the mixture respectively.

### 2.2 Modeling of the adiabatic flame temperature

The adiabatic flame temperature capable of determining the reaction intensity is another fundamental property for premixed flames. It is a measure of the maximum temperature that a combustible mixture can achieve for a given operating condition, which is normally higher in comparison to the flame temperature in the practical combustion systems due to the presence of heat

losses. In general, a high adiabatic flame temperature means a large reaction rate for a reacting mixture [30]. In this work, the adiabatic flame temperature is determined through a gas-phase equilibrium calculation module using ANSYS Chemkin-Pro 2019.

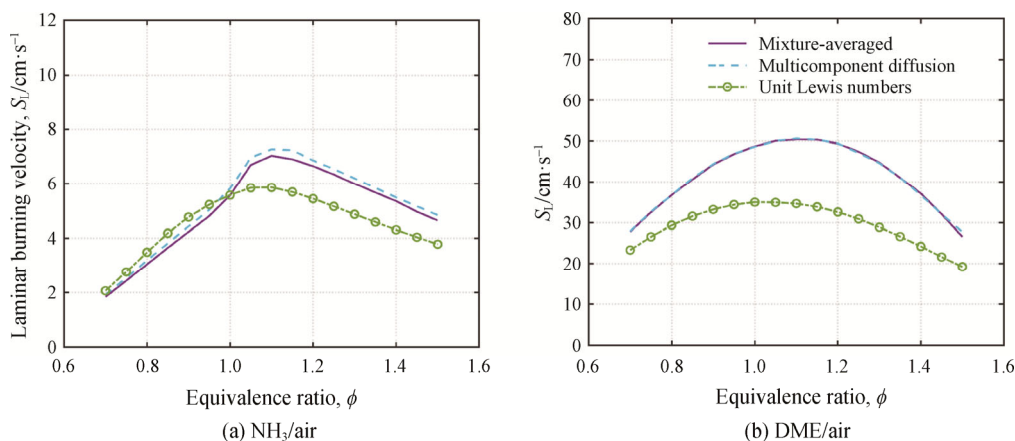
### 2.3 Validating the laminar burning velocity

Since the diffusion transport models play a critical role in the estimated results [31], the impact of these models is first examined. Fig. 1 shows the comparison results of NH<sub>3</sub>/air and DME/air flames considering different diffusion models at the inlet temperature of 300 K. Note that all calculations in this work are performed at a fixed inlet pressure of 101 325 Pa. From Fig. 1(a), it can be seen that under fuel-lean and stoichiometric conditions, the transport models have little influence on the laminar burning velocity of NH<sub>3</sub>/air flames ( $S_{L,NH_3}$ ). In contrast, the calculated  $S_{L,NH_3}$  obtained with these models starts to deviate significantly for fuel-rich conditions. This discrepancy can be explained by examining the effective Lewis number. For a fuel-lean mixture, the deficient component is more heavily weighted [32], so the effective Lewis number is practically that of NH<sub>3</sub>, which

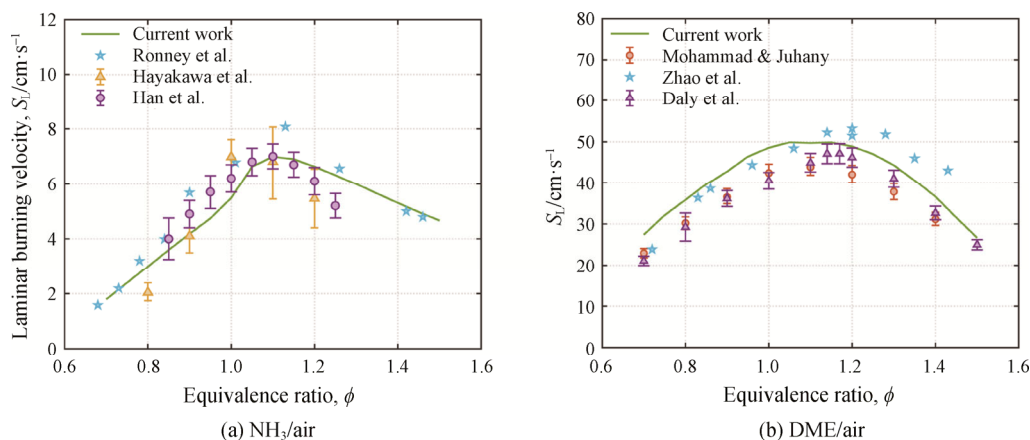
has a Lewis number close to 1.0, leading to the small difference in the laminar burning velocity when using these three models. When it comes to fuel-rich flames, the effective Lewis numbers are far from 1.0, resulting in a large deviation.

Fig. 1(b) shows that good agreement in the calculated laminar burning velocity of DME/air flames ( $S_{L,DME}$ ) can be obtained using the mixture-averaged and multicomponent diffusion models. However, it is worth pointing out that the unit Lewis number model significantly under-predicts the laminar burning velocity. This is because the effective Lewis numbers of DME/air mixture are far from 1.0, independent of the mixture composition. The aforementioned research also revealed that  $S_{L,DME}$  can be well predicted using the mixture-averaged formulation [33]. Accordingly, the mixture-averaged model will be chosen for further parametric studies since it occupies a low computational cost.

To evaluate the feasibility of the reaction mechanism to be used in this work, validation analysis is also conducted. Fig. 2 compares the numerical results using a mixture-average assumption with experimental data



**Fig. 1** The effect of different diffusion models on the laminar burning velocity



**Fig. 2** Variation of experimentally measured and numerically predicted laminar burning velocity of (a) NH<sub>3</sub>/air and (b) DME/air at 298 K

available for both pure  $\text{NH}_3$  and DME/air flames. Experimental data for  $\text{NH}_3$ /air flames using different measurement methods [14, 34, 35] are compared to those numerically obtained, as shown in Fig. 2(a). It can be seen that concerning  $S_{L,\text{NH}_3}$ , the calculated value varies in the range of experimentally measured data. Also, it is interesting to note that the numerically calculated flammability limits of ammonia flames are very close to the theoretically estimated one [29].

Fig. 2(b) compares the experimental and calculated results for DME/air flames [36–38]. The reaction mechanism over-predicts the flame speed of DME/air flames especially under fuel-lean conditions, but the difference between experiments and calculations is negligible. Generally, the calculated flame speed shows a satisfactory agreement with those experimentally measured. Combining the analysis from Fig. 2(a) and (b), it is reasonable to assume that the present chemical reaction mechanism can be applied to describe  $\text{NH}_3$ /DME flame oxidation.

### 3. Results and Discussion

#### 3.1 Temperature dependence of laminar burning velocity

Some previous studies have shown that the combustion process can be enhanced with the implementation of a heat recirculation structure benefiting from the increased inlet enthalpy [39]. In general, increasing the inlet temperature of the mixture is a simple but feasible means to elevate the input enthalpy. This indicates that the inlet temperature of the mixture could also have a strong effect on the laminar burning velocity, in addition to the inlet pressure. To evaluate the extent of such an influence, the estimated laminar burning velocity as a function of the inlet temperature is compared. This is shown in Fig. 3, as the blended ratio is set to 4 different values. It is seen that the laminar burning velocity has a high dependence on the inlet temperature, irrespective of the blended ratio. Increasing the inlet temperature is found to play a positive role in elevating the laminar burning velocity. However, the slope of the laminar burning velocity against the inlet temperature tends to vary depending on the blended ratio. A high blended ratio is found to be associated with a large slope, indicating the more pronounced enhancement effect.

According to the suggestions provided by Egolfopoulos and law [30], the laminar burning flux could be a more proper eigenvalue to express the flame propagation characteristics when compared to the laminar flame speed. Here, the laminar burning flux ( $f^0$ ) is given as

$$f^0 = \rho_u S_L \quad (2)$$

where the superscript 0 represents the initial state, and  $\rho_u$  denotes the density of the unburned mixture.

Fig. 4 shows the variation of the laminar burning flux as a function of the equivalence ratio and blended ratio. It is clear that for a fixed blended ratio, where the freestream density tends to be smaller with elevating inlet temperature, the effect is not sufficiently large to account for the increasing trend of the laminar burning flux, which is actually due to the increasing laminar burning velocity. From an application engineering perspective, the increasing laminar burning flux is desirable as it can enable the combustion system to be operated more stably. These findings further confirm that the blended ratio and freestream temperature play critical roles in the flame propagation, and should be given proper consideration when attempting to enhance  $\text{NH}_3$  combustion.

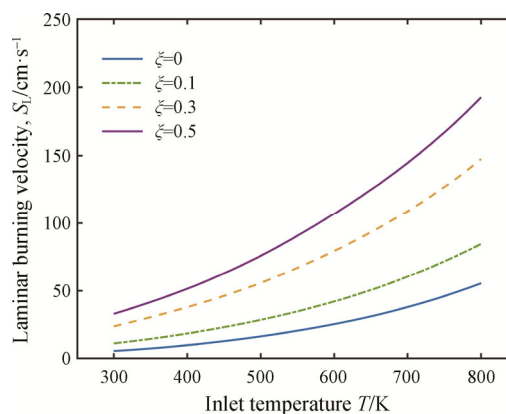


Fig. 3 Variation of the laminar burning velocity as a function of the inlet temperature and blended ratio

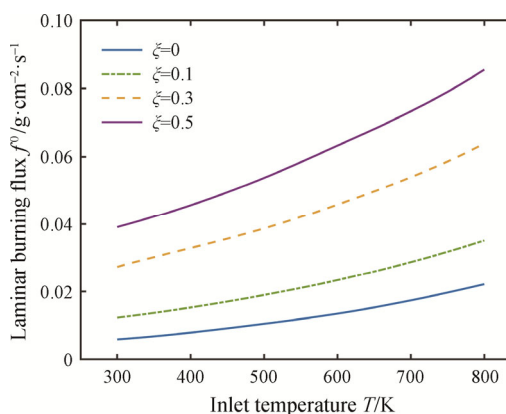


Fig. 4 Comparison of the laminar burning flux as a function of inlet temperature and blended ratio

#### 3.2 Analyses of thermal and diffusion effects

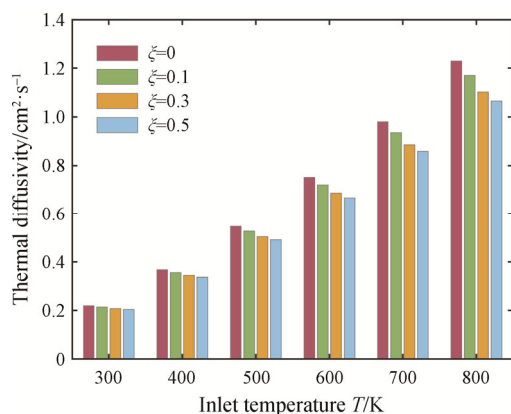
As demonstrated in the previous work [12], the thermal effect plays a key role in promoting the laminar burning velocity when increasing the oxygen content in

the mixture. Therefore, the adiabatic flame temperatures of the  $\text{NH}_3/\text{DME}$  mixture at different inlet temperatures and blended ratios are evaluated to identify its impact on the flame speed, as listed in Table 1. As expected, the adiabatic flame temperature is found to increase significantly with the inlet temperature being elevated. For  $\text{NH}_3/\text{air}$  flames, the adiabatic flame temperature is changed from 2072.0 K at inlet temperature  $T=300$  K to 2358.8 K at inlet temperature  $T=800$  K. Furthermore, it is also noted that increasing the blended ratio gives rise to a higher flame temperature for a given inlet temperature. This is reasonable since DME flames own a higher flame temperature than  $\text{NH}_3$  flames under the same operating conditions.

To gain insight into the relationship between the diffusion effect and the laminar burning velocity, the thermal diffusivity analysis is also implemented. Fig. 5 shows the dependence of the thermal diffusivity on the inlet temperature and blended ratio. It is seen that for a fixed inlet temperature, the thermal diffusivity has a tendency to decrease with increasing the blended ratio. And the increment becomes larger as the inlet temperature is elevated. This indicates that the presence of DME weakens the movement of the mixture. On the contrary, as the mixture composition is fixed, the thermal

**Table 1** Calculated adiabatic flame temperature (K) of  $\text{NH}_3/\text{DME}$  flames at various inlet temperatures and blended ratios

Inlet temperature $T/\text{K}$	Blended ratio ( $\xi$ )			
	0	0.1	0.3	0.5
300	2072.0	2138.7	2207.4	2242.3
400	2131.5	2193.3	2256.5	2288.6
500	2190.1	2246.7	2304.7	2334.1
600	2247.6	2299.1	2351.9	2378.8
700	2304.0	2350.3	2398.1	2422.7
800	2358.8	2400.3	2443.4	2465.7



**Fig. 5** Variation of the thermal diffusivity at various blended ratios and inlet temperatures

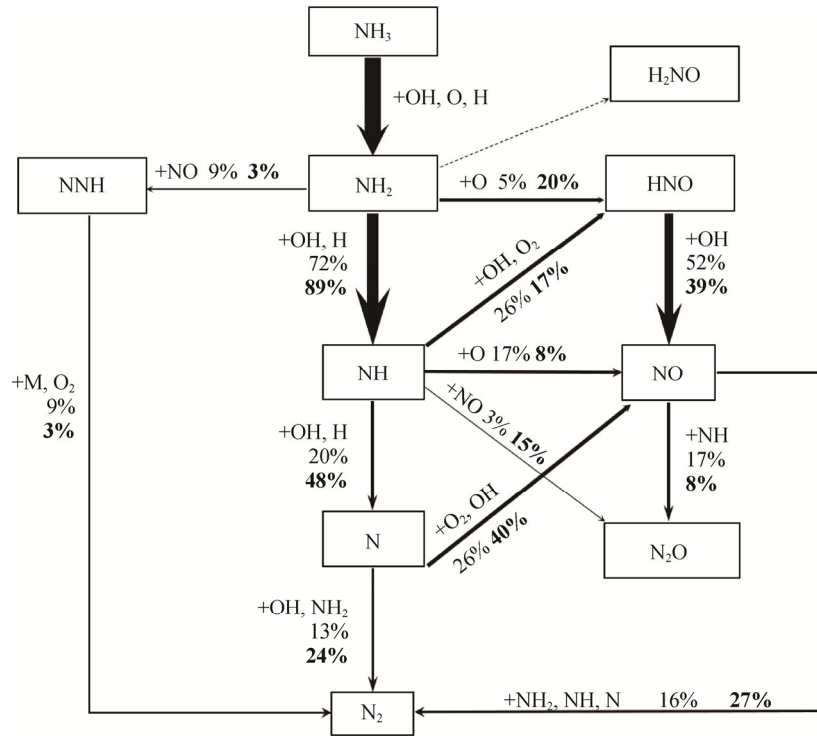
diffusivity is increased remarkably with an increase in the inlet temperature. This means that more heat can be transferred from hot to cold gases for a certain period and contributes to flame propagation.

### 3.3 Kinetic and sensitivity analyses

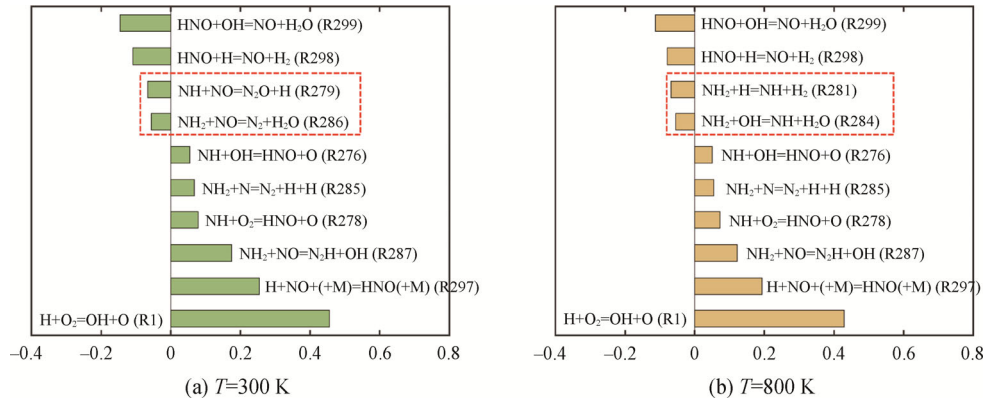
Apart from the two previously-mentioned effects, another effect that is capable of affecting the flame speed in nature is the chemical effect. The kinetic analysis is conducted by first exploring the reaction pathway of  $\text{NH}_3/\text{DME}$  mixtures. The corresponding results are presented in Fig. 6, with  $\xi=0.1$  at  $T=300$  K. The percentage and line thickness correspond to the proportion of N-containing species production via each pathway, which is defined as the integrated rate of species production from a reaction step normalized by the integrated rate of species production from the reactions  $\text{NH}_3 \rightarrow \text{NH}_2$ . These percentages are shown adjacent to the attacking species and each arrow. Also, the dashed lines representing the chemical steps with a percentage less than 0.1% are included for completeness. Fig. 6 shows that  $\text{NH}_3$  is first attacked by active radicals like OH, O, and H to form  $\text{NH}_2$ , and then this radical reacts with others through 3 major pathways to become the final combustion product  $\text{N}_2$ , irrespective of the presence of DME. However, it is worth mentioning that with DME, the reaction intensity of each step varies dramatically. This indicates that the enhanced flame speed of  $\text{NH}_3/\text{air}$  flames with DME added has something to do with the chemical effect.

To further identify the major reaction steps controlling the flame propagation characteristics, a sensitivity analysis is performed. This procedure has the benefit of enabling the sensitivity of species predictions to temperature changes to be evaluated. Fig. 7 presents the normalized sensitivities of the top 10 steps for the laminar burning velocity in stoichiometric  $\text{NH}_3/\text{air}$  flames, as the inlet temperature is varied from 300 K to 800 K. It can be seen that there is no difference in the major reaction steps with the inlet temperature except those two termination reactions highlighted by the red-dotted rectangles. However, it is worth noting that the sensitivity of each step has changed to some extent, especially for the dominant chain branching and terminating steps. The fact that the sensitivities coefficient of the flame speed on the termination steps becomes smaller with the inlet temperature does indicate the progressive importance of the chain-branching reactions. That is, the laminar burning velocity shows a weakening dependence on these reactions as the inlet temperature is elevated.

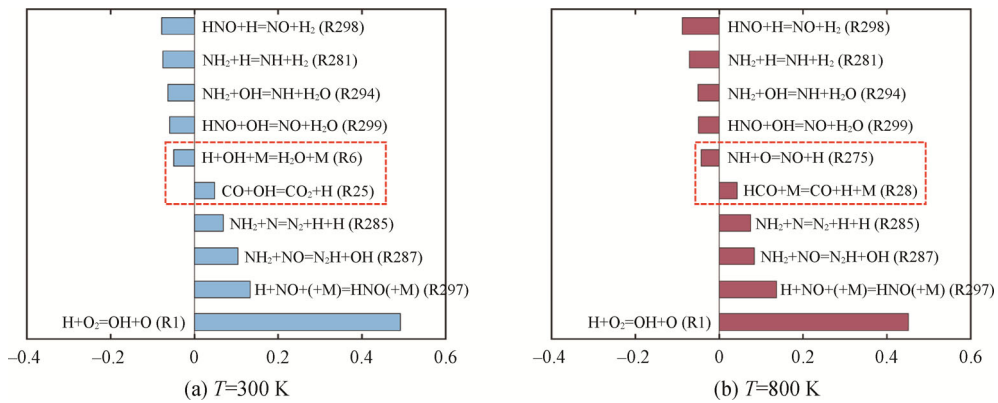
Fig. 8 shows the normalized sensitivities for stoichiometric  $\text{NH}_3/\text{DME}/\text{air}$  flames with  $\xi=0.1$  at 300 K and 800 K. Like  $\text{NH}_3/\text{air}$  flames, varying the inlet temperature leads to two important steps being changed.



**Fig. 6** Reaction pathway for NH<sub>3</sub>/air denoted by regular fonts and NH<sub>3</sub>/DME mixtures with  $\xi=0.1$  represented by bold fronts at  $T=300$  K



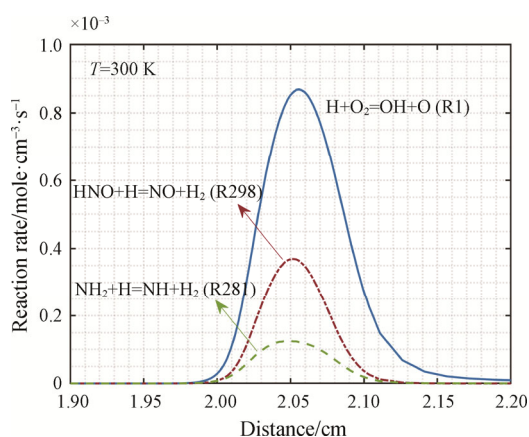
**Fig. 7** Normalized sensitivities on the laminar burning velocity for stoichiometric NH<sub>3</sub>/air flames at  $T=300$  K and  $800$  K



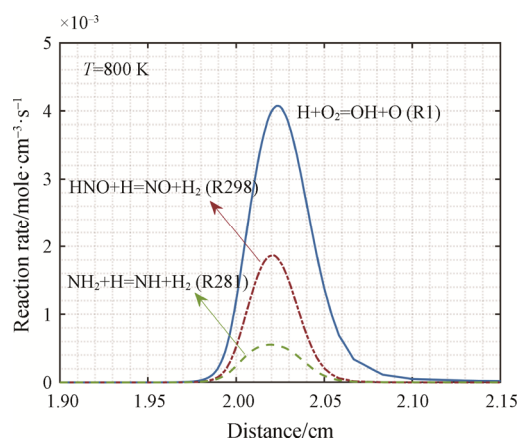
**Fig. 8** Normalized sensitivities on the laminar burning velocity for stoichiometric NH<sub>3</sub>/DME/air flames with  $\xi=0.1$  at  $T=300$  K and  $800$  K

However, the relative importance of the termination steps over chain-branching steps has changed little with the inlet temperature, and so are the sensitivities of these steps. This indicates that the chemical effect does not play a dominant role in the flame speed variation with the inlet temperature. Another observation by comparing Figs. 7(a) with 8(a) is that in the presence of DME, the termination reactions have changed considerably except for the most dominant chain-branching one. Meanwhile, it is worth noting that although there is only 10% of DME in the total fuel, the reaction R25 becomes one of the most important chain-branching steps affecting the laminar burning velocity, as shown in Fig. 8(a) and (b), suggesting the progressive significance of reactions involving C-atom.

The reaction rate variations of the three major dominant chain reactions for the stoichiometric  $\text{NH}_3/\text{DME}$  flame as the initial temperature is varied from 300 K to 800 K are shown in Figs. 9 and 10 respectively. It can be seen that there is a slight difference in the importance of the competition between the branching and termination reactions as the inlet temperature is elevated, which is a manifestation of the weak influence of the chemical effect. This is in satisfactory agreement with those results illustrated in Figs. 7 and 8. Meanwhile, it is interesting to mention that in an elevated inlet temperature condition, the reaction rate of all the chain-branching and terminating steps are increased dramatically. This is mainly because of the higher adiabatic flame temperature. By comparing and analyzing the adiabatic flame temperature in Table 1 and sensitivity coefficients in Figs. 7–10, it is concluded that the increased laminar burning velocity in  $\text{NH}_3/\text{DME}/\text{air}$  flames with elevating the inlet temperature is primarily due to the thermal effect resulting from the higher adiabatic flame temperature and diffusion effect, rather



**Fig. 9** Profiles of the numerically calculated rate of production of several important branching and terminating reactions for the binary mixture flames with  $\xi=0.1$  at  $T=300$  K



**Fig. 10** Profiles of the numerically calculated rate of production of several important branching and terminating reactions for the binary mixture flames with  $\xi=0.1$  at  $T=800$  K

than the chemical effect. This is similar to the situations in oxygen-enriched combustion [12]. On the contrary, the flame speed enhancement in the  $\text{NH}_3/\text{air}$  mixture with DME added is a result of thermal and chemical effects.

#### 4. Conclusions

In this work, the laminar burning velocity of  $\text{NH}_3/\text{DME}/\text{air}$  flames spanning a wide range of the inlet temperature and blended ratio is numerically evaluated using ANSYS Chemkin-Pro 2019. The laminar burning velocity is observed to have a monotonic dependence on the inlet temperature and the blended ratio. Increasing these two parameters contributes to a higher laminar flame speed. It is quite beneficial to the flammability limit and combustion process in practical combustion systems. To reveal the mechanisms underlying the enhancement phenomenon, detailed analyses are performed in terms of the thermal and diffusion, followed by the reaction pathway and sensitivity studies.

Regarding the enhanced flame speed with DME and the elevated inlet temperature, it is found that both these two implementations give rise to a higher adiabatic flame temperature, which is beneficial to the overall reaction rate. Meanwhile, the thermal diffusivity is shown to vary considerably with the inlet temperature, but increased slightly with the blended ratio. Finally, detailed kinetic and sensitivities analyses reveal that the presence of DME has significantly changed the major dominant branching and terminating steps and their relative importance. However, such influence is quite little, when elevating the inlet temperature is concerned. Accordingly, it can be concluded that the increased laminar burning velocity in  $\text{NH}_3/\text{DME}/\text{air}$  flames with elevating the inlet temperature is primarily due to the thermal effect resulting from the higher adiabatic flame temperature and

the diffusion effect, rather than the chemical effect. On the contrary, the flame speed enhancement in the NH<sub>3</sub>/air mixture with DME added is the synergistic result of thermal and chemical effects. In general, this work sheds light on how the thermal, diffusion, and chemical effects affect the laminar burning velocity of a combustible mixture, when different enhancement strategies are applied.

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