NUMERICAL STUDY ON COMBUSTION AND EMISSION OF NH_3/CH_4 MIXTURES UNDER GAS TURBINE CONDITIONS: EFFECTS OF PRESSURE AND HYDROGEN

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To improve the combustion performance of ammonia/ methane mixtures, this study, based on the Chemkin PREMIX model and the Okafor reaction mechanism, analyzes the effects of varying hydrogen content, initial pressure, and equivalence ratio on laminar burning velocity and associated radical concentrations of ammonia/ methane mixtures. The study investigates the promotional effect of hydrogen addition on the combustion reaction chain and quantitatively analyzes the linear relationship between laminar burning velocity and H+NH_2 radical concentrations. The results show that hydrogen addition significantly enhances the combustion speed, with a peak at an equivalence ratio of 1.05–1.1, while laminar burning velocity decreases with increasing pressure. The increase in hydrogen content promotes the formation of key radicals such as H, OH, and NH_2, Also, the study reveals a strong linear correlation between laminar burning velocity and the concentration of max (H + NH_2) radicals, with this relationship being significantly influenced by the pressure change.

Key words*: Laminar burning velocity; radical concentration; carbon emissions; ammonia; hydrogen; methane emission;*

1. Introduction

The ongoing exploitation and utilization of fossil fuels have significantly contributed to the intensification of the global greenhouse effect[1, 2]。In response, the United Nations Framework Convention on Climate Change calls for a reduction in fossil fuel consumption[3], while China's "2030 Carbon Peak Action Plan" outlines three core objectives: increasing the share of non-fossil energy in total consumption, improving energy efficiency, and reducing the carbon intensity of emissions[4]. In this context, research on renewable low-carbon fuels has garnered significant attention, sparking a new wave of scientific inquiry. A critical challenge in the development of these fuels lies in achieving their efficient utilization in practical applications.

Gas turbines, as key power devices, offer substantial potential for application, particularly in the search for alternative fuels to replace natural gas and reduce carbon emissions. Natural gas, primarily composed of methane, is commonly used in gas turbines; however, methane combustion can lead to carbon emissions[5]. Therefore, exploring the co-combustion of low-carbon, active fuels with natural gas is of paramount importance[6, 7].

Ammonia and hydrogen are considered ideal low-carbon fuel alternatives[8, 9]. However, ammonia combustion faces significant challenges, particularly in low-temperature environments, hindering its practical application[10]. The addition of hydrogen significantly enhances combustion reactivity and efficiency but introduces the risk of flashback. Introducing ammonia can effectively mitigate this issue, providing a promising strategy for improving the performance of low-carbon fuels in practical applications.

Current studies on the fundamental combustion characteristics of ammonia-hydrogen-methane mixtures primarily focus on binary blends, such as ammonia/hydrogen[11-13]and ammonia/methane[14, 15], while research on ternary ammonia-hydrogen-methane mixtures remains limited. Among the few available studies, Wang et al.[16]conducted turbulent combustion experiments in a turbulent combustion chamber under conditions of ϕ = 0.6–0.91 and T = 373 K, examining the effects of pressure (1 and 5 atm) and Lewis number on turbulent flame speeds. Berwal et al.[17, 18]investigated the impact of ammonia content on the laminar burning velocity (LBV) of hydrogen-methane mixtures under high-temperature (T = 300–750 K) and high-pressure (1–5 atm) conditions, reporting a decrease in LBV with increasing ammonia content. Additionally, Mashruk et al.[9]used a generic swirl burner to study the optimal mixture ratio of ammonia/hydrogen/methane under atmospheric pressure and T = 288 K with ϕ = 1.2, concluding that a mixture ratio of 55/25/20% exhibited the best performance, characterized by low NO emissions and stable reactions. Bayramoğlu et al.[19]further explored the influence of ammonia content on NOx emissions from ammonia/hydrogen/methane combustion, demonstrating that co-combustion of hydrogen, ammonia, and traditional fuels effectively reduces emissions while improving combustion performance.

Pressure has been a key focus in gas turbine applications[16, 17, 20, 21], with most studies conducted at pressures ranging from 1 to 5 atm. However, modern heavy-duty F-class gas turbines typically operate at pressures as high as 18 atm. Despite this, studies on combustion and emission characteristics under a broader pressure range remain scarce. Furthermore, prior research has primarily focused on NOx emissions, with limited attention to CO emissions.

This study builds upon the optimal ammonia/hydrogen/methane mixture ratio of 55/25/20% identified by Mashruk et al.[9]and simplifies it to a 2/1/1 ratio. A detailed analysis was conducted to investigate the effect of a wide pressure range (3–15 atm) on LBV and radical concentrations under conditions of ϕ = 0.7–1.3 and T = 423 K. Using MATLAB, the linear relationship between LBV and key radicals was quantitatively examined to elucidate the variations in LBV with changes in pressure and radical concentrations. Given the critical role of radicals (H, O, and OH) and pressure in influencing LBV[17, 22], , this study further explored the impact of pressure on these radicals and combustion emissions. The results revealed a quantitative relationship between LBV and the maximum concentration of H+NH_2 radicals. These findings provide a theoretical foundation for understanding the combustion and emission mechanisms of ammonia/hydrogen/methane mixtures under practical gas turbine operating conditions.

2. Numerical analysis

2.1. Chemical kinetic analysis

The fundamental combustion characteristics of ammonia have been extensively studied, driving the development and optimization of chemical kinetic mechanisms for ammonia combustion. While numerous studies have focused on ammonia/hydrogen and ammonia/methane combustion, relatively few mechanisms have been developed specifically for ammonia/hydrogen/methane mixtures. Four reaction mechanisms—Okafor[23], Shrestha[24], GRI[25] and CEU-NH_3[26]—are available for the numerical simulation of ammonia/hydrogen/methane combustion. The details of these mechanisms, including the number of species and reactions, are summarized in Table 1.

The Shrestha mechanism[24], developed based on laminar burning velocity experimental data from Lhuillier et al.[27], focuses on ammonia oxidation and NOx formation but tends to overestimate burning velocities. Wang et al.[26]introduced the CEU-NH_3 mechanism, tailored for NH_3/CH_4/CH_3OH/C_2H_5OH mixtures, which showed improved predictive capabilities for ammonia-methane mixtures. However, the GRI-Mech 3.0[25]mechanism, widely used for methane-ammonia flames, underpredicts burning velocities due to its limited representation of NO reduction pathways. To address these limitations, Okafor et al.[23]integrated the GRI-Mech 3.0[25]and Tian[28]mechanisms to develop a new ammonia-methane mechanism. higher pressures enhance the reaction

As shown in Fig. 1, this study validates the performance of the four mechanisms using experimental data from ammonia-hydrogen (P = 1–5 atm, T = 298 K), ammonia-methane (P = 1–5 atm, T = 298 K), and ternary ammonia-hydrogen-methane mixtures (P = 5 atm, T = 300–750 K). The results indicate that the Shrestha mechanism overestimates the laminar burning velocities for ternary mixtures, while the GRI-Mech 3.0 mechanism underestimates them. The CEU-NH3 mechanism accurately predicts burning velocities for ammonia-hydrogen and ammonia-methane mixtures but overestimates them for lean ternary mixtures. In contrast, the Okafor mechanism demonstrates excellent predictive accuracy for laminar burning speeds across all conditions, aligning closely with experimental results.

Therefore, the Okafor mechanism was selected for this study to investigate the combustion characteristics of ammonia/hydrogen/methane mixtures.

Tab.1 Verify the required mechanisms and characteristics

Reaction Mechanism	Species	Reactions	Publication Date
Okafor [23]	59	356	2019
Shrestha [24]	125	1099	2021
GRI [25]	53	325	1999
CEU-NH_3 [26]	91	444	2021

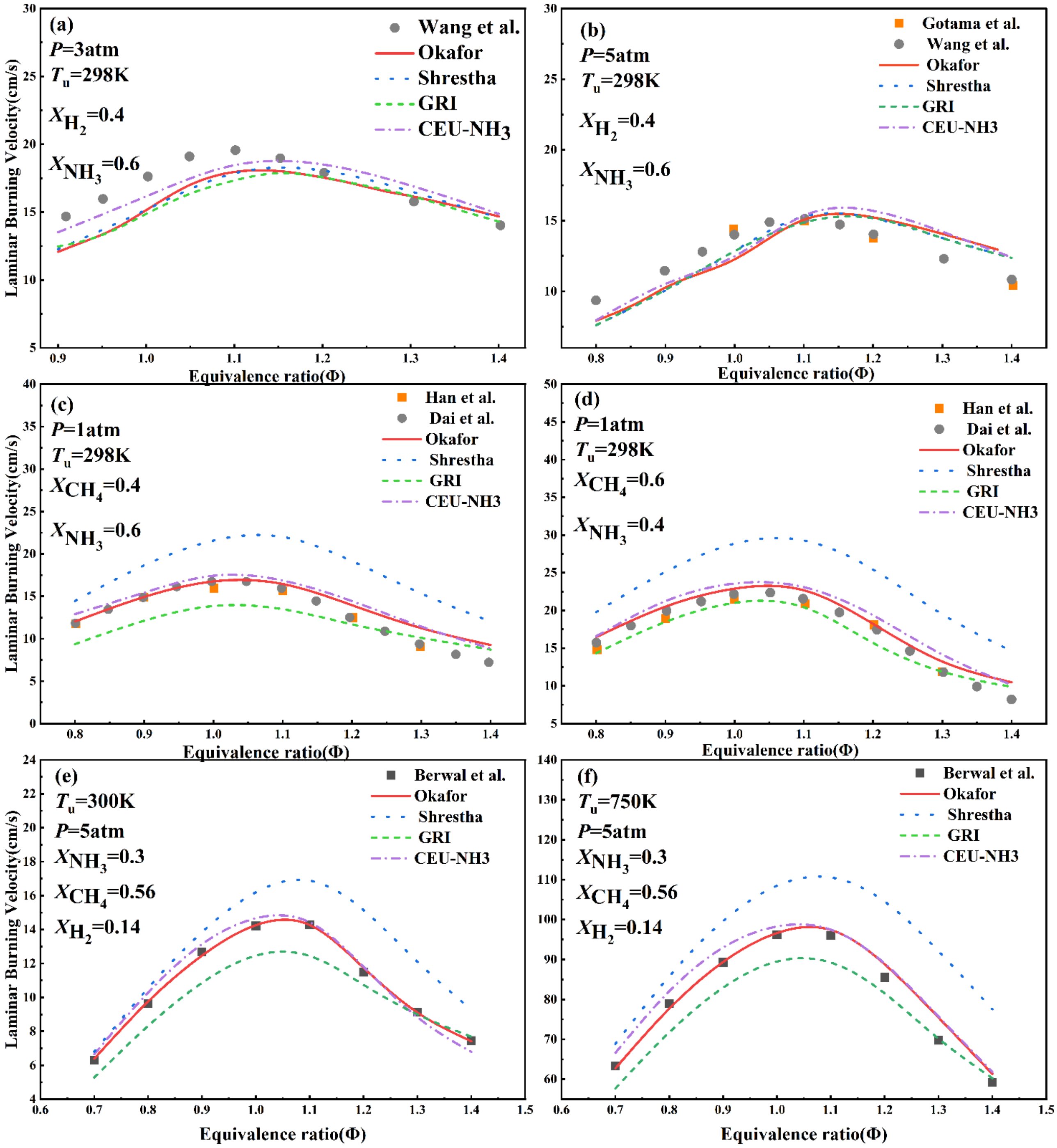


Fig. 1. Experimental and simulated laminar flame speeds (symbols: experimental data from Wang[29], Gotama[30], Han[31], Dai[22], Berwal[17]; lines: simulated data)

2.2. Numerical simulation

The combustion of ammonia/hydrogen/methane mixtures was simulated using the one-dimensional freely propagating PREMIX model in Chemkin. The flame structure during combustion is depicted in Fig. 2, where the laminar premixed flame is divided into a preheat zone and a reaction zone. In the preheat zone, the mixture undergoes continuous heating, while in the reaction zone, combustion and oxidation reactions occur. The "distance" in this context refers to the propagation distance of the laminar premixed flame.

For this study, the computational domain spanned 0–10 cm, and the grid was refined with a total of 2,000 grid points, including 200 adaptive grids. Grid refinement was based on the convergence criteria of GRAD = 0.1 and CURV = 0.1. To reflect actual gas turbine operating conditions, the simulations accounted for the Soret effect, with an initial temperature set at 423 K. Six pressure levels were considered, ranging from 3 atm to 15 atm. The specific simulation parameters are summarized in Table 2.

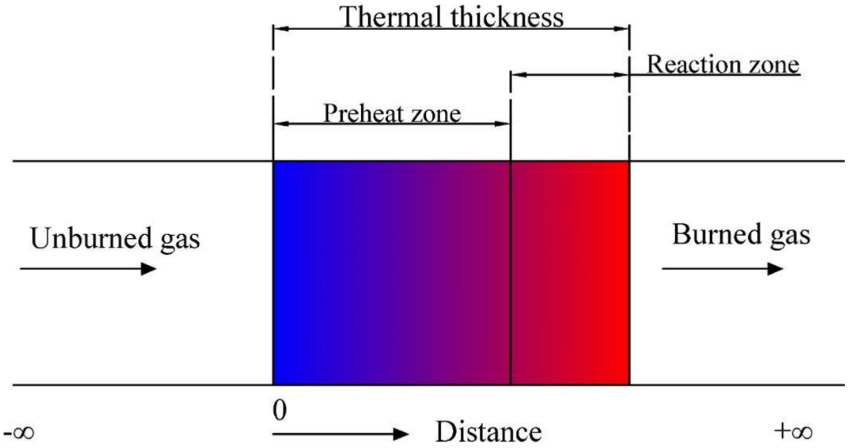


Fig. 2. Schematic diagram of laminar premixed flame

Tab.2 Initial conditions for numerical calculations

Case	Fuel Composition			Pressure	Equivalence Ratio	Temperature
	NH_3	H_2	CH_4			
1	0.5	0.25	0.25	3atm	0.7-1.3	423K
2				5atm		
3				8atm		
4				10atm		
5				13atm		
6				15atm		
7	0.5	0	0.5	5atm		
8	0.5	0.1	0.4			
9	0.5	0.25	0.25			
10	0.5	0.4	0.1			
11	0.5	0.5	0			

3. Numerical analysis

3.1. Effect of pressure and hydrogen content on laminar burning velocity

The efficiency of gas turbines depends heavily on the combustion chamber operating under high temperatures and pressures[32]. Therefore, investigating the laminar burning velocity (LBV) of ammonia/hydrogen/methane fuels under such conditions is crucial for understanding their combustion characteristics. This section examines the effects of pressure and hydrogen content on LBV to provide insights into the combustion behavior of these ternary fuel mixtures.

Fig. 3 illustrates the variation of LBV with equivalence ratio (ϕ), initial pressure (P = 3–15 atm), and hydrogen content (X_{H_2} = 0–0.5). In all cases, LBV initially increases with the equivalence ratio, reaching a peak at ϕ = 1.05–1.1, and then decreases. The peak laminar burning velocity (LBV) decreases with increasing pressure, as elevated pressures not only accelerate the reaction $H+O_2 \rightarrow O+OH$ (R39), promoting the consumption of H radicals, but also shift the chemical equilibrium toward the formation of more stable intermediates such as H_2O and CO_2. Consequently, the concentrations of reactive radicals like H, O, and OH are reduced, effectively suppressing chain-branching reactions and thereby decreasing the LBV[33]. Fig. 3(b) highlights that LBV increases significantly with higher hydrogen content, also peaking at ϕ = 1.05–1.1 before declining. Compared to a pressure increase from 3 atm to 5 atm, which results in a 26% increase in peak LBV, a rise in hydrogen content from X_{H_2} = 0.4 to 0.5 yields a more pronounced 32% increase. This indicates that hydrogen content has a stronger effect on LBV than pressure, with pressure's impact on LBV ultimately linked to the influence of H radicals.

To further understand the flame structure of the ternary mixtures, Fig. 4 shows the impact of hydrogen content (X_{H_2} = 0.1–0.4) and pressure (P = 5–13 atm) on the reaction rates of key pathways involved in the production and consumption of H radicals. Three critical reactions were selected for analysis: R85 ($OH + H_2 \rightleftharpoons H + H_2O$) and R3 ($O + H_2 \rightleftharpoons H + OH$), which are the primary sources of H radicals, and R39 ($H + O_2 \rightleftharpoons O + OH$), which predominantly consumes H radicals.

With increasing pressure and hydrogen content, the production and consumption rates of H radicals both increase. Notably, the yield of H radicals exhibits a marked increase as hydrogen content rises. When hydrogen content increases from X_{H_2} = 0.1 to 0.4, the rate of reaction R39 doubles, while the rates of R85 and R3 increase by a factor of approximately one. These findings highlight the significant role of hydrogen content in enhancing the production of H radicals, particularly under elevated temperature conditions.

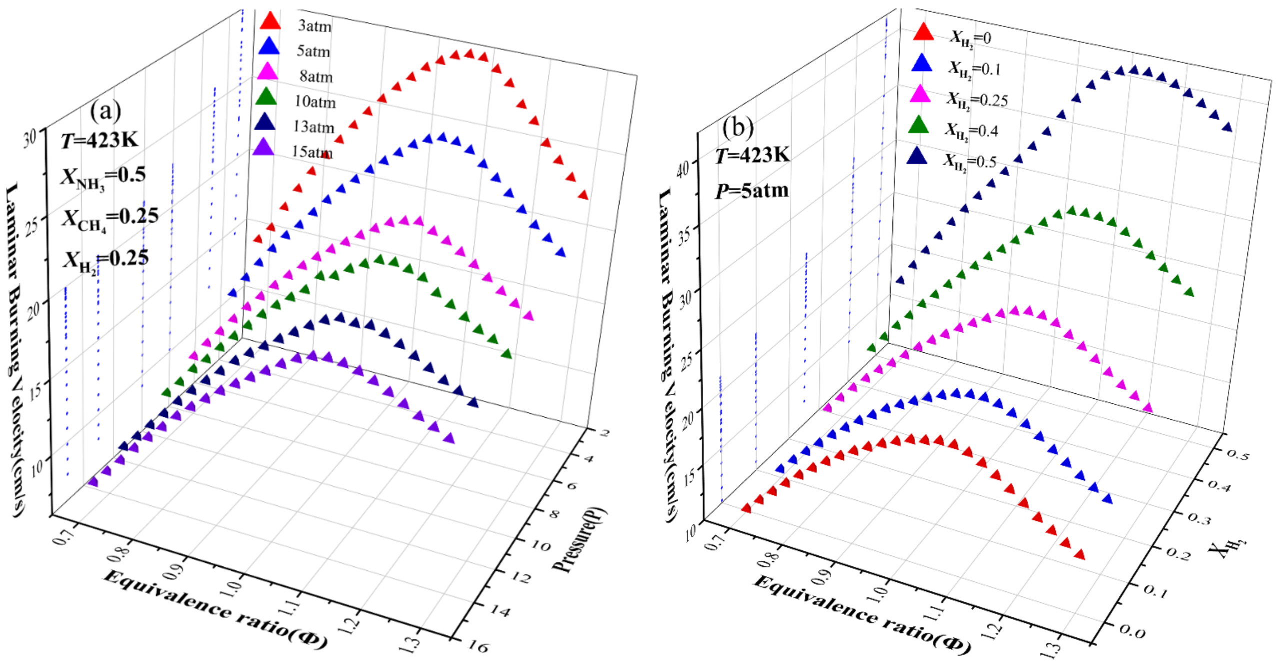


Fig. 3. Relationship between laminar burning velocity and equivalence ratio under different pressures and hydrogen contents

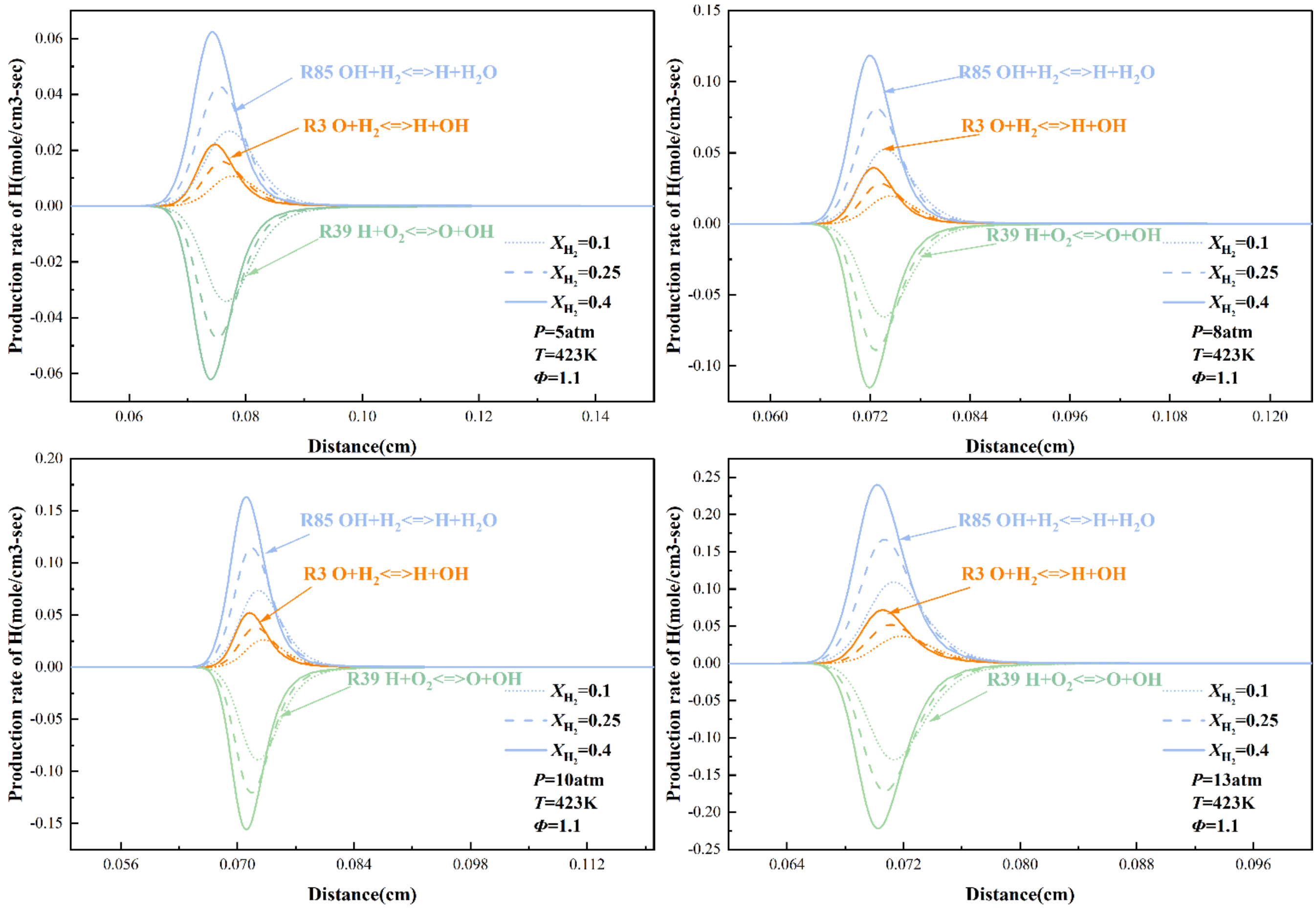


Fig. 4. Sensitivity analysis related to H under different pressures (T=423K, ϕ=1.1，X_{H2}=0.1，0.25，0.4）

3.2. Relationship between laminar combustion velocity, pressure, and free radicals

Previous studies have demonstrated that the production and consumption of active radicals significantly influence laminar burning velocity (LBV)[34-36]. Liu et al.[37]showed that the LBV of C_3H_8-O_2-N_2 and C_3H_8-O_3-CO_2 mixtures is proportional to the maximum concentration of (H + OH + O). Similarly, Wang et al. [38] established a linear relationship between LBV and the maximum concentration of (H + OH + CH_3) in CH_4/DME/H_2 premixed flames using a heat flux burner. Our previous studies indicated that the LBV of ammonia-hydrogen and ammonia-hydrogen-carbon monoxide mixtures correlates linearly with the maximum concentration of (H + NH_2) radicals[39-41]. These findings collectively suggest that LBV is closely associated with radical concentrations.

To validate this relationship for ternary ammonia/hydrogen/methane mixtures, simulations were conducted at T = 423 K and P = 5 atm. The LBV and the maximum concentrations of key radicals (H, O, OH, and NH_2) were recorded. Fig. 5 illustrates the variation of these parameters with equivalence ratio (ϕ) under different pressures. The results reveal that increasing pressure suppresses the maximum concentrations of H, O, OH, and NH_2, while equivalence ratio has differential effects on these radicals.

On the lean side ($\phi < 1.0$), the maximum concentration of OH increases rapidly with the rise in equivalence ratio, reaching its peak at $\phi = 0.95$, and then decreases sharply on the rich side. The trend for O is similar to that of OH. This behavior is primarily attributed to the higher O_2 concentrations in the lean region, where reaction R39 ($H + O_2 \rightarrow O + OH$) becomes more pronounced, leading to increased production of O and OH radicals. As ϕ increases toward the rich region, the dissociation of NH_3 releases more H_2, which subsequently promotes reactions R3 ($O + H_2 \rightarrow H + OH$) and R85 ($OH + H_2 \rightarrow H + H_2O$), consuming O and OH radicals and resulting in their decline.

The variations of max(H), max(H + NH_2), and LBV with equivalence ratio exhibit consistent trends across different pressures, with all peaking at $\phi = 1.1$. Both H and NH_2 are identified as key radicals influencing LBV. A clear linear relationship is observed between LBV and the maximum concentrations of H and (H + NH_2). However, while max(H) aligns well with LBV under lean conditions, the correlation deteriorates under rich conditions. Overall, LBV shows the strongest and most consistent linear correlation with max(H + NH_2), as depicted in Figs. 6 and 3(a). Notably, this linear relationship evolves systematically with changes in pressure.

As illustrated in Fig. 7, the progression of the fuel mixture from the unburned region to the combustion zone involves complex chemical reactions. Hydrogen content significantly affects the generation and distribution of key radicals such as H, OH, and NH_2. At zero hydrogen content, the peak concentrations of H and OH radicals are relatively low. However, as hydrogen content increases, the production of these radicals occurs earlier, and their peak concentrations rise substantially, indicating that hydrogen enhances the intensity of combustion reactions and the activity of the radical chain reaction.

Under stoichiometric conditions (ϕ = 1.0), increasing hydrogen content markedly elevates the peak concentrations of H, OH, and NH_2 radicals, facilitating more complete combustion. In contrast, under rich conditions (ϕ = 1.3), the concentration of NH_2 radicals is significantly higher compared to stoichiometric conditions, particularly at elevated hydrogen levels. This behavior is attributed to the increased dissociation of ammonia and more active chain reactions in the rich combustion environment, which promote NH_2 formation. Conversely, the concentration of OH radicals decreases under rich conditions, consistent with previous studies[39-41]. This reduction in OH levels in the rich regime contributes to lower NO_x emissions. Overall, the addition of hydrogen accelerates the radical chain reaction, leading to more efficient combustion.

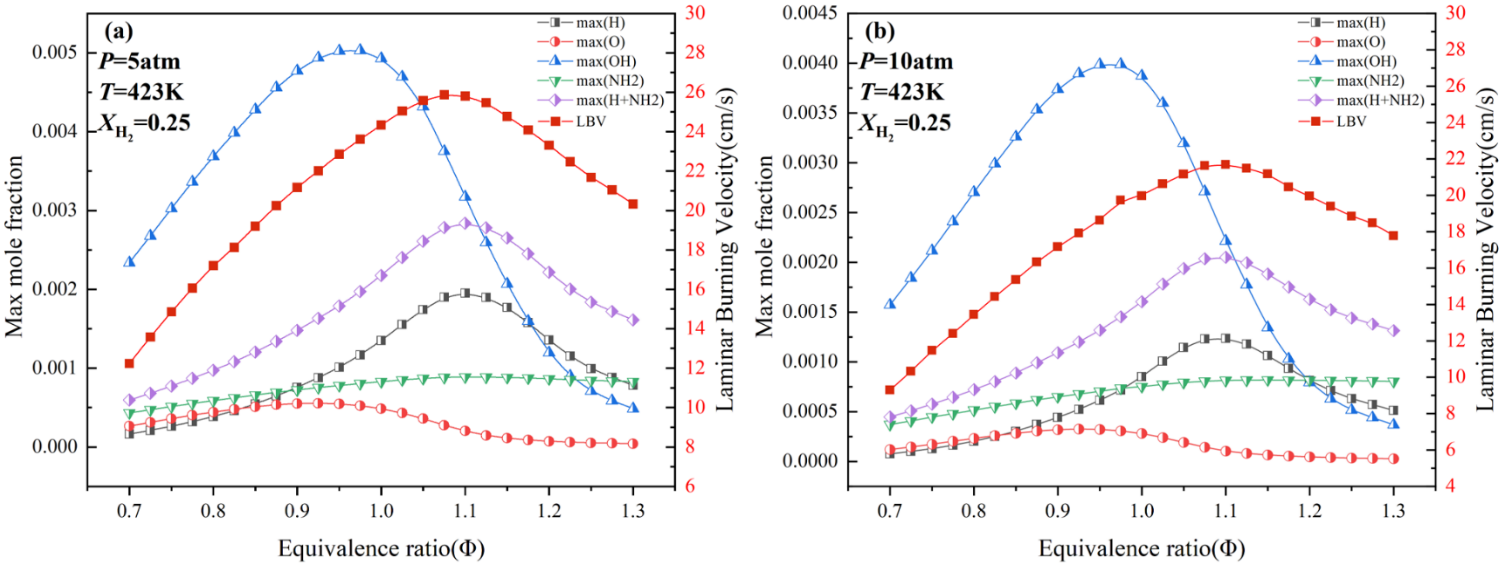


Fig. 5. The influence of pressure on max (O), max (H), max (OH), max (NH2), max (H+NH2), and LBV (T=423K, X_{H2}=0.25）

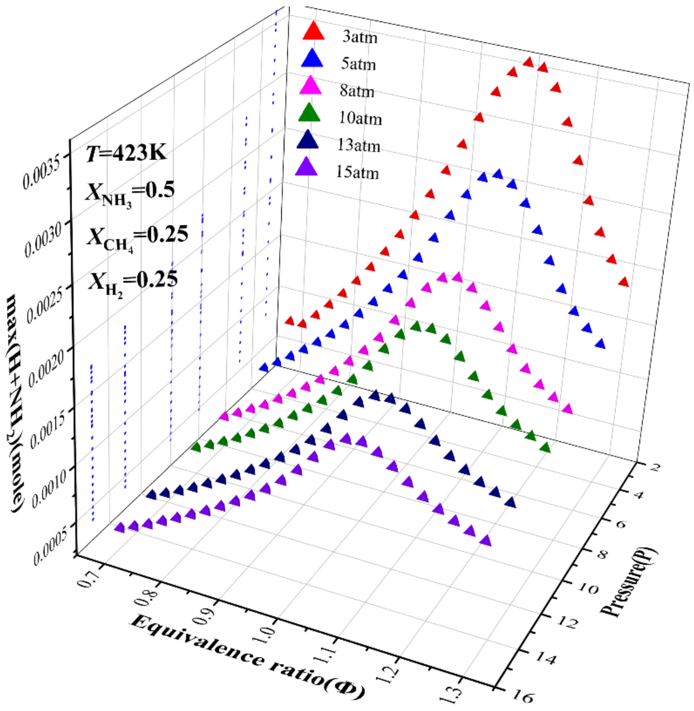


Fig. 6. The relationship between max (H+NH_2) and equivalence ratio under different pressures

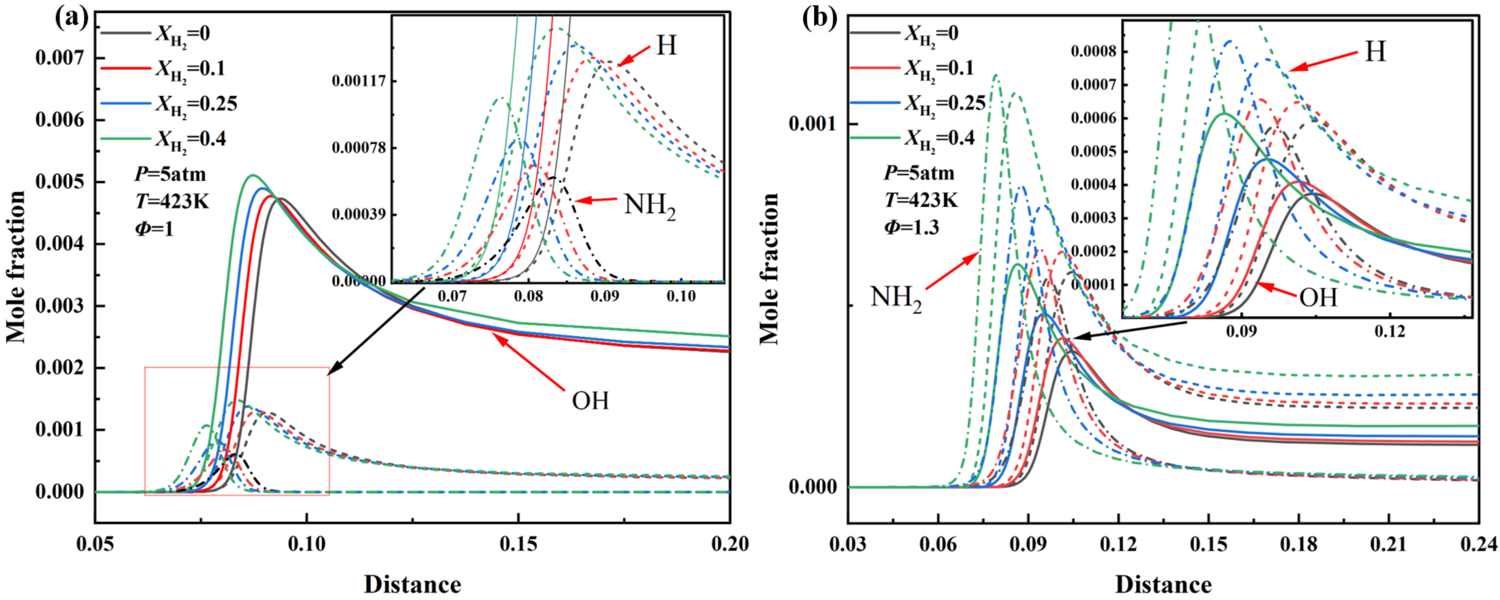


Fig. 7. Mole fractions of free radicals (H, NH$_2$, OH) with different hydrogen contents and equivalence ratios (ϕ=1-1.3， X_{H2}=0-0.4）

Fig. 8 illustrates the linear relationship between max(H + NH$_2$) and laminar burning velocity (LBV) under various pressures (3–15 atm), equivalence ratios (Φ = 0.7–1.3), and hydrogen contents (X_{H_2} = 0.1–0.4) at T = 423 K. The results indicate that this linear relationship remains consistent regardless of equivalence ratio or fuel composition. This finding aligns with Cai et al.[39] who observed a similar independence of this relationship from hydrogen content and equivalence ratio in ammonia-syngas mixtures. However, Wang et al.[38]reported that this relationship varies with hydrogen content in CH$_4$/DME/H$_2$ blends, suggesting that fuel composition plays a significant role in determining the correlation between LBV and other variables.

Table 3 presents the slope (p1), intercept (p2), and correlation coefficient (R^2) of the linear fits at different pressures. It is evident that p_1 increases with pressure, while p_2 decreases, indicating a positive correlation between slope and pressure and a negative correlation between intercept and pressure.

Further analysis of p1 and p2 against pressure (Fig. 9) reveals a linear dependence within the range of 3–10 atm. Accordingly, the LBV can be expressed as a function of pressure and max(H + NH$_2$):

$$S_{L,P}=(528.84P+2694.66)\max(H+NH_2)-1.07P+17.85$$

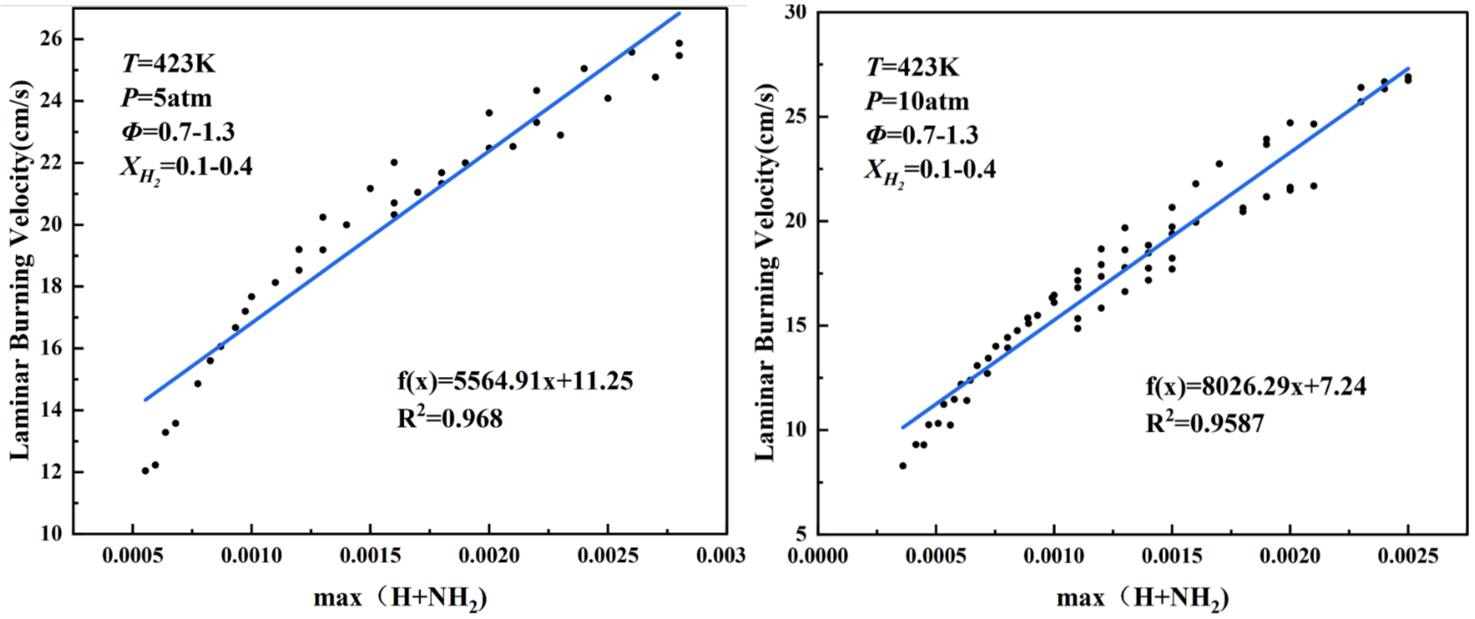
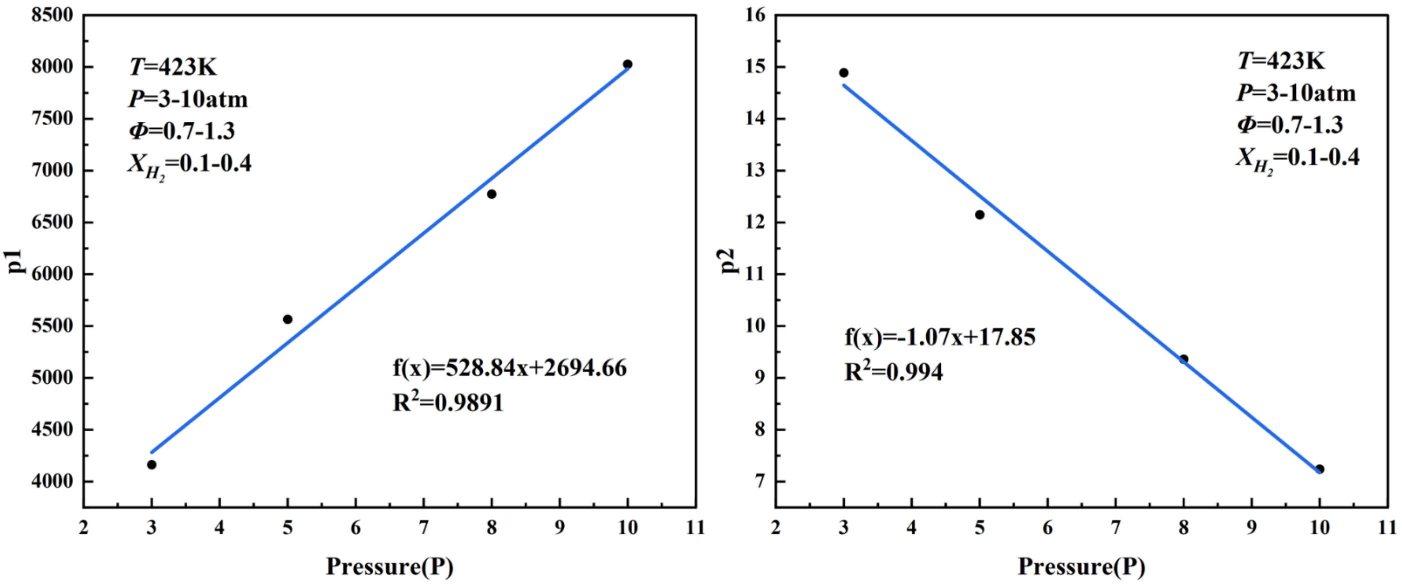


Fig. 8 The functional relationship between laminar combustion velocity and max ($H+NH_2$) at P=5atm and P=10atm

Tab.3 Linear correlation coefficient between max ($H+NH_2$) and lbv under different pressures

Case	pressure	Slope (p1)	Intercept (p2)	R^2
1	3	4162.83	14.89	0.9692
2	5	5564.91	12.15	0.968
3	8	6774.34	9.36	0.9751
4	10	8026.29	7.24	0.9587

Fig. 9 The linear relationship between slope p1 and intercept p2 and pressure



3.3. Sensitivity analysis

The effects of hydrogen content and initial pressure on max($H + NH_2$) and laminar burning velocity (LBV) were further analyzed under the given conditions ($\Phi = 1.1$, $T = 423$ K, $X_{H_2} = 0.1–0.4$). Figs. 10 and 11 illustrate the sensitivity of key reactions to variations in pressure and hydrogen content.

Notably, the reaction R39 ($H + O_2 \rightleftharpoons O + OH$) consistently emerges as the dominant pathway for H radical consumption. This reaction plays a critical role in laminar premixed combustion, acting as the initiating step in chain branching reactions[33, 42]. As hydrogen content increases, the dominant sensitivity of most reactions to radical dynamics decreases. This observation aligns with the findings of Shu et al.[43], Wang et al.[29], and Masoumi et al.[44]. Among these, the sensitivity of R39 shows the most pronounced reduction, attributed to the involvement of H_2, which facilitates the formation of active radicals such as O, H, and OH[33].However, under high-pressure conditions, the sensitivity of R39 increases, promoting a higher consumption rate of H radicals and potentially reducing LBV. This pressure effect intensifies the recombination of radicals, thereby suppressing chain branching reactions and limiting flame propagation. Additionally, reaction R53 ($H + CH_3(+M) \rightleftharpoons CH_4(+M)$) plays a significant role in moderating radical sensitivity by suppressing chain reactions.

In contrast to the hydrogen content's enhancing effect on radical sensitivity, increasing the initial pressure demonstrates a strong inhibitory influence on LBV. Notably, the sensitivity of R39 increases significantly with rising pressure, resulting in elevated consumption of H radicals. This phenomenon corresponds closely with the trends in H radical production rates shown in Fig. 4.

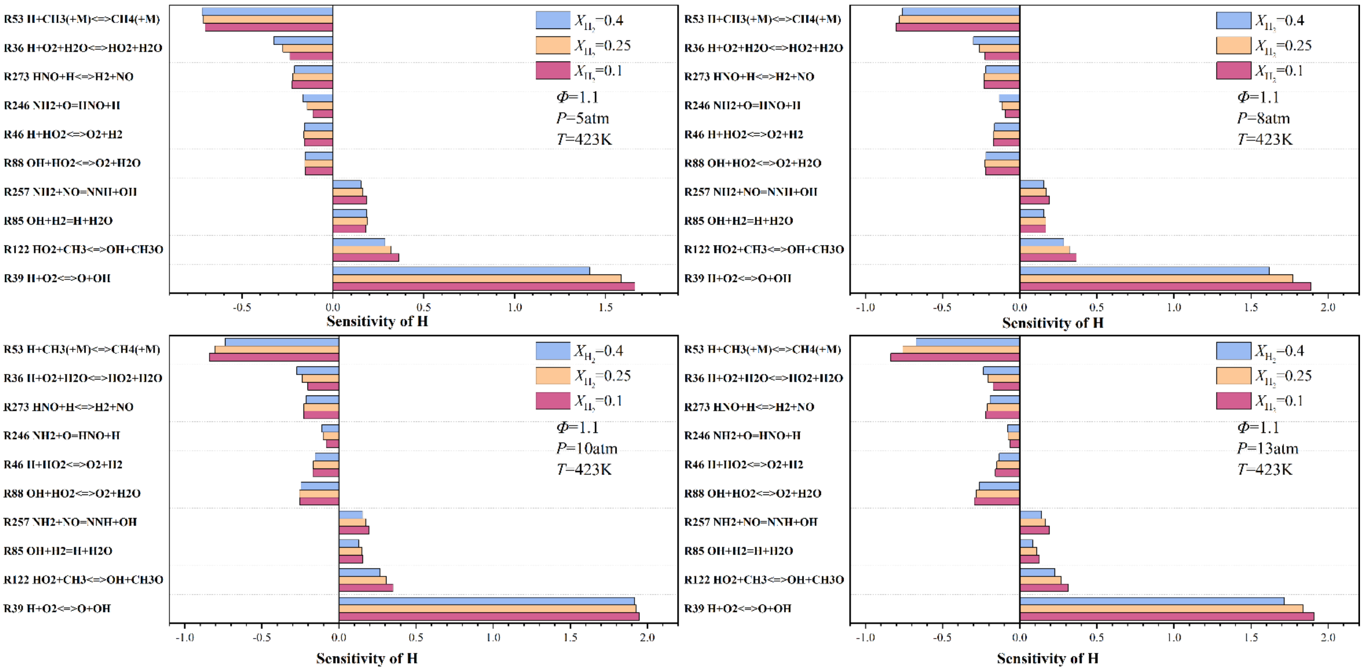


Fig. 10 Sensitivity analysis of H under different pressures (T=423K, Φ=1.1, X_{H2}=0.1-0.4）

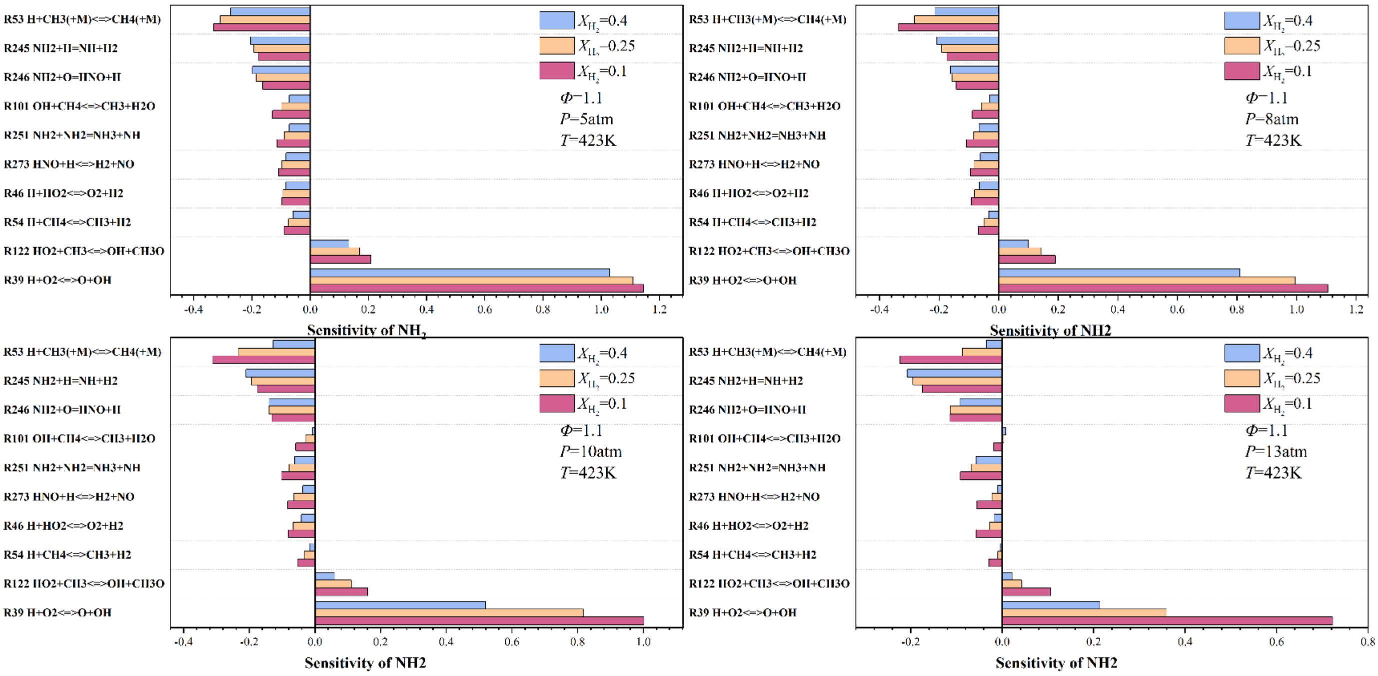


Fig. 11 Sensitivity analysis of NH_2 under different pressures (T=423K, Φ=1.1, X_{H2}=0.1-0.4)

Conclusion

This study systematically investigates the laminar premixed combustion characteristics of ammonia/hydrogen/methane fuel mixtures under varying pressure, hydrogen content, and equivalence ratio using a one-dimensional freely propagating flame model. The main findings are as follows:

1. The increase in hydrogen content significantly enhances the laminar burning velocity (LBV), while an increase in pressure leads to a decrease in LBV, particularly under rich combustion conditions. The maximum LBV is observed at equivalence ratios between 1.05 and 1.1.

2. A strong linear correlation exists between LBV and the maximum (H + NH_2) radical concentration, which is influenced by pressure. The slope and intercept of this relationship can be quantified as functions of pressure.

3. The reaction R39 ($H + O_2 \rightarrow O + OH$) is identified as the primary pathway for the consumption of H radicals, playing a critical role in chain branching reactions. The addition of hydrogen reduces the sensitivity of R39, while enhancing the formation of active radicals.

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