

Effect of Hydrogen Addition on Soot Formation and Emission in Acetylene Laminar Diffusion Flame

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Cite This: *ACS Omega* 2023, 8, 24893–24900

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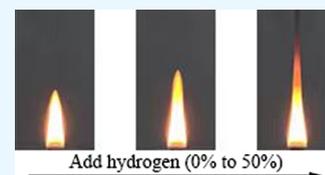


Article Recommendations



Supporting Information

ABSTRACT: Hydrogen (H₂) has been regarded as a highly competitive alternative fuel that does not produce CO₂ and soot during combustion. There are few studies of cofiring H₂ with hydrocarbons. Meanwhile, the effect of hydrogen addition on soot formation and emission is questionable. In this study, the effect of H₂ addition with varying ratios (between 0 and 50% by molar fraction while the remainder is nitrogen) on soot formation in acetylene (50% by molar fraction) laminar diffusion flames was experimentally and numerically investigated. Results show that with H₂ addition, the flame height increases and the temperature decreases. The total soot emission and the primary particle size both increase with H₂ addition. The addition of H₂ promotes soot formation. In addition, the soot oxidation is weakened due to the lower flame temperature. Chemical kinetic analysis shows that the concentrations of Al, H, and H₂O increase with H₂ addition, which is consistent with the experimental results. According to the HACA reaction, the increase of the molar fraction of Al and H radicals could promote PAH growth and soot formation.



1. INTRODUCTION

Soot^{1–3} is produced by incomplete combustion and insufficient oxidation of fuel.⁴ The soot particulate emission from the combustion of hydrocarbon fuels would cause serious environmental pollution and thus endanger human health.^{5,6} Soot particles are considered to be a second contributor to global warming.⁷ With the proposal of new carbon emission regulations, it is distinctly important to improve conventional combustion processes and reduce soot formation and emission.⁸ Many studies have been devoted to reducing the soot emission.⁹ In the past several years, lots of studies have been focused on the investigation of the soot formation process of some practical fuels, such as natural gas, gasoline, kerosene, diesel, etc.¹⁰

In the past few years, hydrogen (H₂) has received unprecedented attention as a green fuel that does not produce CO₂ and soot during combustion. However, due to safety factors related to the issues of storage and transportation of hydrogen, as well as the potential of hydrogen explosions,¹¹ the application of hydrogen as a fuel has not yet been met with great success. Previous studies mainly focused on the fundamental combustion characteristics of cofiring H₂ with hydrocarbons.^{12–15} Some important combustion characteristics such as flame temperature, swirl intensity, and fuel mixture compressibility¹⁶ are reported to be adjusted with the addition of H₂. Compared with traditional hydrocarbon fuels, hydrogen has a lower combustion limit and a higher heating value. Hydrogen is an important carbon-free fuel, cofiring H₂ with hydrocarbons could achieve a better combustion process, and it can improve the combustion efficiency and reduce the pollutant emission, such as CO₂ and NO_x.¹⁷ Previous research showed that these fuels can improve combustion and pollutant emission performance, such as combustion ignitability,

stability, and NO_x.^{18–21} Hydrogen as supplementary fuel could increase the flame speed and expand the lean flammability limits.^{22,23} Therefore, H₂ is often used to be mixed with hydrocarbon fuels or as an additive to hydrocarbon fuels, such as CNG and H₂ mixture in spark ignition engines,²⁴ non-premixed H₂ methane (CH₄)/N₂ oxygen (O₂) flame,²⁵ and CH₄–O₂ injection flame added with H₂.²⁶

In fact, the soot formation and emission of cofiring H₂ with hydrocarbons are rarely studied. Park and Haynes et al. investigated the effect of hydrogen on the soot formation through experiments. Pandey et al.²⁷ have studied the effects of hydrogen addition on soot formation in acetylene-air laminar diffusion flames. Their results also demonstrate that the soot volume fraction decreases with hydrogen addition. Haynes et al.²⁸ have studied the influence of gaseous additives on the formation of soot in premixed flames. Their results show that the soot yield is not significantly affected by additions of up to 3% H₂ to the unburned gases.

Wei et al.²⁹ have studied the effects of hydrogen addition on soot particle size distribution functions in laminar premixed flame. Their results show that molar concentrations of important soot precursor species, such as C₂H₂ and PAHs, are inhibited by the addition of hydrogen, and hydrogen is an effective additive for reducing soot formation. Yen et al.³⁰ have studied the influence of hydrogen and nitrogen addition on

Received: February 23, 2023

Accepted: June 16, 2023

Published: July 5, 2023



soot formation in laminar ethylene jet diffusion flame. When the molar fraction of hydrogen reaches 40%, the total loading of soot is decreased by 30%, and when the molar fraction of nitrogen reaches 40%, the total loading of soot is decreased by 70%. Park et al.³¹ have investigated the effects of hydrogen addition on soot formation and oxidation in laminar premixed C₂H₂/air flames. Their findings indicate that the addition of hydrogen can slow the oxidation rate of acetylene but has less effect on inhibiting soot. All the above studies are simulation research.

The effect of hydrogen addition on soot formation and emission in hydrocarbons is questionable. There are contradictions between the previous research conclusions. In addition, most of the relevant research is only numerical simulation. This work focuses on the effect of H₂ addition on soot formation in C₂H₂ laminar diffusion flame at atmospheric pressure. Acetylene is selected as the reference fuel because it is one of the common hydrocarbon fuels and one of the important species for soot formation. The influence of hydrogen addition with different ratios (between 0 and 50% by molar fraction while the remainder is nitrogen, N₂) on soot formation and emission in acetylene (50% by molar fraction) laminar diffusion flame are investigated experimentally and numerically. In addition, the influence of hydrogen addition on the concentration of key species in the soot formation process is studied by chemical kinetics analysis.

2. EXPERIMENTAL SETUP

2.1. Laminar Diffusion Flame Burner and Flame Description. In this work, the laminar diffusion flame of acetylene doped with hydrogen and nitrogen was adopted as the research object to perform the experiment. Figure 1 shows

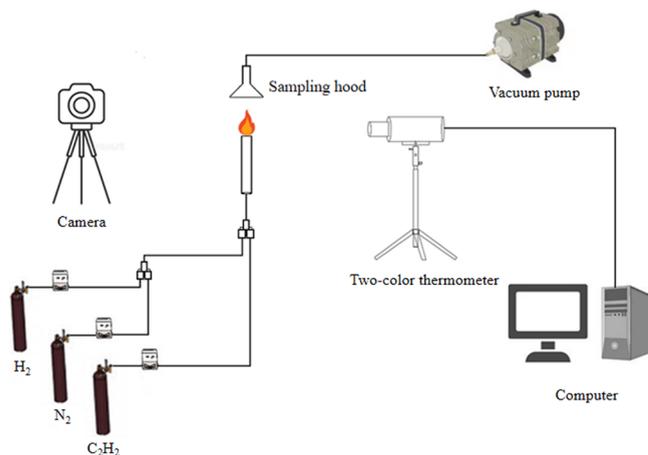


Figure 1. Schematic of the experimental setup.

the schematic of the experimental system including the burner, gas supply system, sampling system, image acquisition system, and temperature measurement system. The burner consisted of a stainless-steel tube (SUS316), with an inner diameter of 2 mm and an outer diameter of 4 mm, which was fixed on an optical platform. The mixed gases were supplied to the burner through a network system consisting of compressed gas cylinders, mass flow meters, valves, and pipes. The combustion products were removed by a blower located approximately 500 mm above the burner outlet. Three mass flow controllers (Alicat) with an accuracy of 0.5% were used to control the flow

rates of acetylene (purity $\geq 98\%$), hydrogen (purity $\geq 99.9\%$), and nitrogen (purity $\geq 99.9\%$). All experiments in the present study were conducted at atmospheric pressure and room temperature (298 K).

Eleven gaseous fuel mixtures were explored in this research. The flow rate of acetylene was set to 50 mL/min (298 K, 1 atm). As shown in Table 1, the concentration of C₂H₂ was

Table 1. Parameter Setting of Fuel Composition

fuel	X_{H_2}	$F_{C_2H_2}$ (mL/min)	F_{H_2} (mL/min)	F_{N_2} (mL/min)
H0N50	0	50	0	50
H5N45	0.05	50	5	45
H10N40	0.1	50	10	40
H15N35	0.15	50	15	35
H20N30	0.2	50	20	30
H25N25	0.25	50	25	25
H30N20	0.3	50	30	20
H35N15	0.35	50	35	15
H40N10	0.4	50	40	10
H45N5	0.45	50	45	5
H50N0	0.5	50	50	0

fixed at 50% and the N₂ diluent was substituted with H₂. The molar fraction of hydrogen in the fuel, X_{H_2} , is defined as the ratio of the hydrogen flow rates to the total fuel flow rates. To facilitate the description of these cases, fuels at X_{H_2} of 0 to 0.5 are marked as H0N50 to H50N0, respectively. The principle for determining the fuel flow rate is to ensure that all 11 fuels have the same carbon atom flow rates.

2.2. Temperature Measurement and Image Acquisition. In this work, the temperature profiles along the flame centerline were measured by a two-color thermometer (produced by advanced-energy, Onyx-S2C, the temperature measurement range is 700 to 3000 °C). The flame temperature measurement points are shown in Figure 2. The lower, middle,



Figure 2. Temperature measurement points.

and upper of the flame were selected as temperature measurement points, because these three positions could express the temperature change trend of the whole flame. According to Guo et al.,³² the thermometer was aimed at the position of interest, and then the temperature within 1.5 mm of the location was tested in a period of 2 ms, last for half a minute. The flame temperature at each position was measured

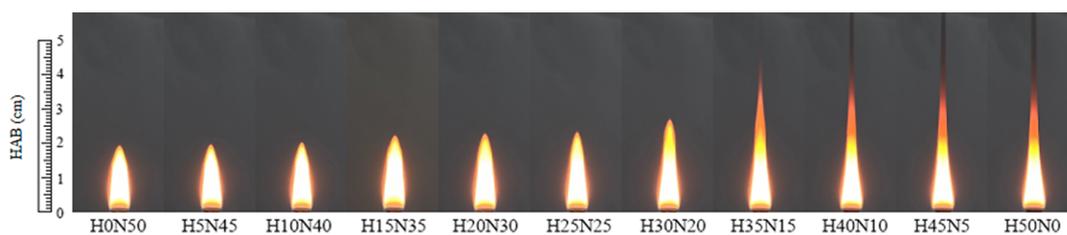


Figure 3. Visible flame appearances with various levels of H₂ addition.

at least three times, and the average value was taken to ensure accuracy. During the flame temperature measurement, the uncertainty of the flame temperature is about ± 0.2 °C.

The flame images were obtained by camera acquisition, and accurate flame image was obtained by manual focusing. A professional image processing software Adobe Premiere was utilized to process flame images. For each flame image, the photographs were repeated at least five times to select the best image and to eliminate the instability of the experiment.

2.3. Soot Sampling. The sampling system is composed of a vacuum pump, a vacuum tube, a sampling hood, and glass fiber filter paper. The vacuum pump was connected with the sampling hood through a vacuum tube, and then the glass fiber filter paper was fixed on the sampling hood. The glass fiber filter paper with a pore size of 1 μm was selected, and the back of the filter paper was pumped to a negative pressure by a vacuum pump to ensure that the soot could be completely collected. The soot emission by the flame was collected by the sampling system within 1 min, and then the total mass of soot particles was measured by electronic balance (artorius, BSA2245-CW). The uncertainty of the electronic balance is approximately ± 0.1 mg. For each case, measurements were repeated at least three times for average to ensure accuracy.

2.4. Transmission Electron Microscopy (TEM) Analysis. A 200 kV field emission transmission electron microscope (FEI Talos 200S) was used to obtain low-resolution and high-resolution bright field images of soot particles. In this study, soot particles were photographed at 36 kX, 74 kX, 150 kX, 310 kX, and 630 kX magnification. For each soot sample, 20 images were taken to ensure that the image was clear and the data were accurate. In addition, the samples were observed within 24 h after sampling to avoid oxidation of soot particles. The average soot particle diameter was obtained by professional image processing software Digital Micrograph.

2.5. Chemical Kinetic Modeling. To gain insight into the PAH formation process of the acetylene-hydrogen flame, a H₂-C₂H₂-PAH chemical mechanism was constructed to explain the concentrations of key species for soot formation at different levels of H₂ addition.

The comprehensive reaction mechanism developed by GRI 3.0 and Frenklach³³ was referenced as the fundamental mechanism. Both detailed kinetic mechanisms have been validated extensively against experimental results and are consistent with the conclusions of this research. The diffusion flame model in CANTERA was used for numerical simulation, the calculation was completed by Python 3.8.2. The chemical kinetic mechanism in CANTERA format can be found in the [Supplementary Materials](#).

3. RESULTS AND DISCUSSION

3.1. Visible Flame Appearance and Temperature.

Figure 3 shows the steady-state flame photographs of the

H0N50 to H50N0 flames. The visible flame height is defined as the vertical distance between the burner exit and the position where soot particles are entirely burnt out.³⁴ The visible flame heights of H0N50 to H50N0 flame increase monotonically with H₂ addition. The heights of H0N50 to H30N20 flames are 19.52, 19.83, 20.61, 22.41, 23.28, 23.90, and 27.71 mm, respectively. From H35N15 to H50N0 flames, the soot breaks through the flame, forming soot wings. This causes the visible flame height to be not measured accurately. Since nitrogen does not participate in the combustion process, the addition of nitrogen does not increase the consumption of oxygen, whereas hydrogen will participate in the combustion process and increase the consumption of oxygen. Therefore, the stoichiometric air-fuel ratio increases with hydrogen addition, which may be one reason for the increase of the flame height. Another possible reason for the increase of the visible flame height is that H₂ has a longer ignition delay time and lower burning velocity than C₂H₂. The orange luminescence region in the upper of flame gradually expands with H₂ addition and soot begins to be emitted when $X_{\text{H}_2} = 35\%$, which indicates that H₂ dramatically promotes the soot formation and emission. The results indicate that H₂ addition could promote soot formation and emission.

The temperature profiles along the flame centerline are shown in Figure 4. Different from other studies, the flame temperature tends to decrease gradually with the addition of H₂. The maximum temperatures of H0N50 to H50N0 flames are 1897.2, 1895.2, 1889.1, 1888, 1883.4, 1879.6, 1876.7, 1875.3, 1872.2, 1867.4, and 1860.8 °C, respectively. With the H₂ addition, the maximum flame temperature decreases

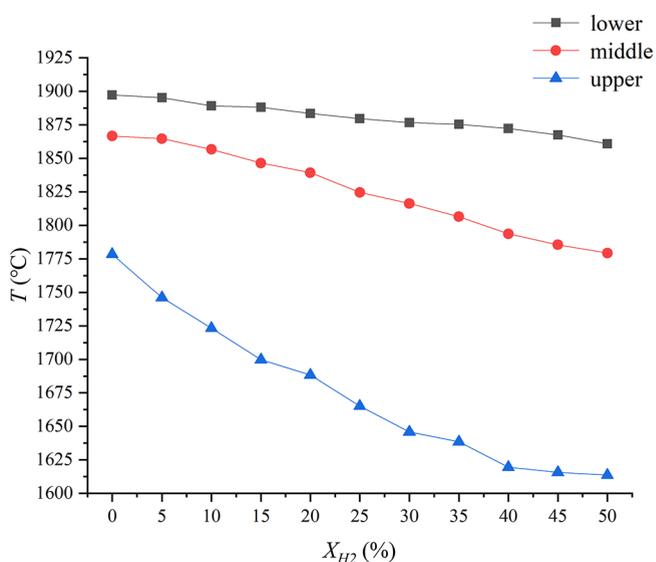


Figure 4. Measured flame temperature profiles along the centerline.

slightly. When $X_{H_2} = 50\%$, the maximum flame temperature decreases by $36.4\text{ }^\circ\text{C}$. It can be seen from Figure 4 that the temperature in the upper of the flame drops faster than that in the middle and lower of the flame. The same observation is also obtained by Liu et al.³⁵ by addition of H_2 to the methane/air flame. On the one hand, H_2O is the principal product of hydrogen combustion. The presence of more H_2O can reduce the flame temperature. In addition, the specific heat of H_2 is much higher than that of N_2 , and is an order of magnitude higher, which is probably the main reason why the flame temperature decreases with the H_2 addition. The adiabatic combustion temperatures of hydrogen and acetylene were obtained through thermodynamic calculations. The adiabatic combustion temperature of hydrogen is lower than that of acetylene, and the flame temperature will decrease with hydrogen addition. Another possible reason is that the presence of H_2O would enhance radiative losses reducing the flame temperature. In addition, the increased soot formation by adding H_2 should increase the soot radiative losses, which may lead to a slight decrease in temperature. The reduction of flame temperature could slow down the reaction rate, such as the growth of PAHs, soot inception and surface growth.³⁶ The lower flame temperature is more conducive to the soot formation and emission. In addition, H_2O has slightly effect on soot formation, but the presence of H_2O could weaken the oxidation reactivity of soot and reduce flame temperature.³⁷ The lower flame temperature could weaken the oxidation process of soot in the flame, thus leading to an increase in soot emission.

The above results indicate that H_2 addition could promote soot formation and emission in acetylene laminar diffusion flame. To our knowledge, this is different from previous studies. Wei and Yen et al. have found that hydrogen addition could reduce soot formation in acetylene flame.^{29,30} Pandey and Park et al. have found that hydrogen addition has little effect on soot formation in acetylene flame.^{27,31}

3.2. Effect of Hydrogen Addition on Soot Formation and Emission. Soot formation and emission is an extremely complex physicochemical process. A general consensus has been reached on soot formation, which includes fuel pyrolysis, incipient ring formation, PAH growth, particle inception, surface reaction, particle-particle interactions, oxidation, and fragmentation.^{3,38} The purpose of this section is to explore the effect of hydrogen addition on soot formation and emission in acetylene laminar diffusion flame.

As shown in Figure 5a, when $X_{H_2} = 0$ to 25% , regardless of the sampling time, soot cannot be collected. This indicates that the flame has no obvious soot emission. When $X_{H_2} = 30$ to 50% , there is a linear relationship between the mass of collected soot and the sampling time. This indicates that the flame has obvious soot emission. Figure 5b shows the formation and emission rates of soot at different times. The trend of soot formation and emission rate is the same for different sampling times. The experimental result is consistent with visible flame appearances in Figure 3. The orange-yellow region in the flame gradually increases with the hydrogen addition, which indicates that soot formation increases. However, not all of the soot that is formed may be oxidized following this pathway through high-temperature oxidizing regions. When $X_{H_2} = 30$ to 50% , soot wings appear, with the soot breaking through the flame.

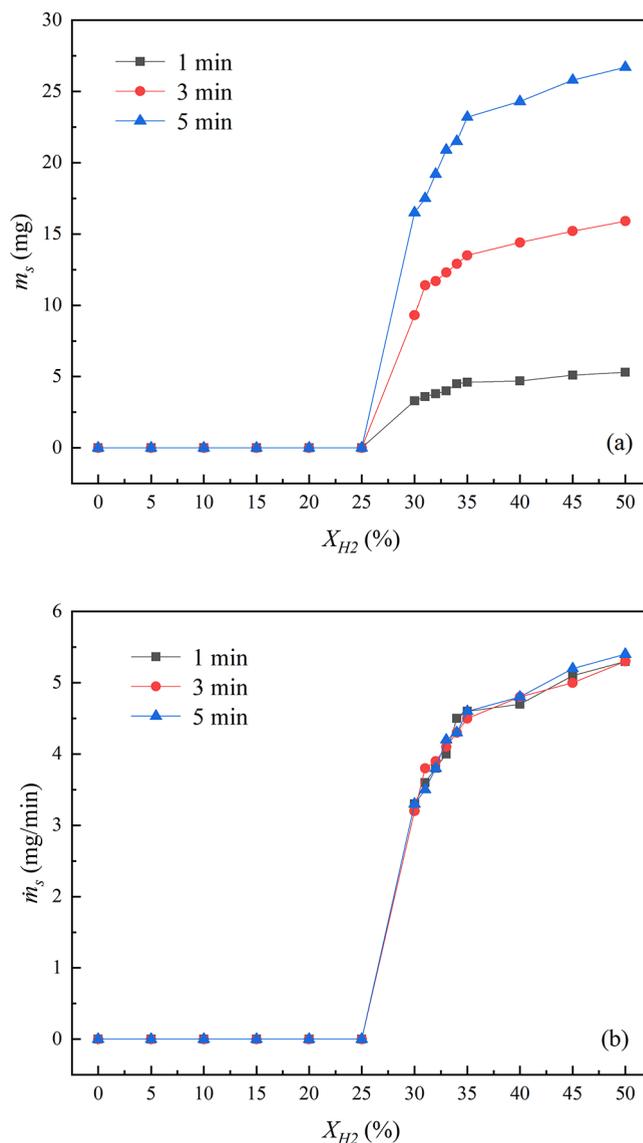


Figure 5. Effect of H_2 addition on soot formation and emission.

3.3. TEM Analysis. It is generally believed that the size of soot particles increases through surface reaction growth, and most of the soot mass comes from surface growth. Thus, the average particle size of soot could reflect the surface growth rate of soot particles to a certain extent. In addition, the aggregation of soot particles is also an important factor in the soot formation and emission. Figure 6 shows the average particle size of soot from H35N15 to H50N0 flames. The morphology of corresponding soot particles is shown in Figure 7. The primary soot particle size of H35N15, H40N10, H45N5, and H50N0 flames is 20.1, 23.4, 27.7, and 33.2 nm, respectively. The particle size of soot increases almost linearly with the hydrogen addition. Moreover, as shown in Figure 7, the aggregation of soot particles also increases with the H_2 addition. The results further suggest that hydrogen is not an effective soot inhibitor. On the contrary, hydrogen could increase the soot emission by promoting soot particle surface growth and particle aggregation.

3.4. Chemical Kinetic Analysis. The experimental results show that the addition of H_2 promotes the soot formation and emission, but the underlying cause is not clear. In order to

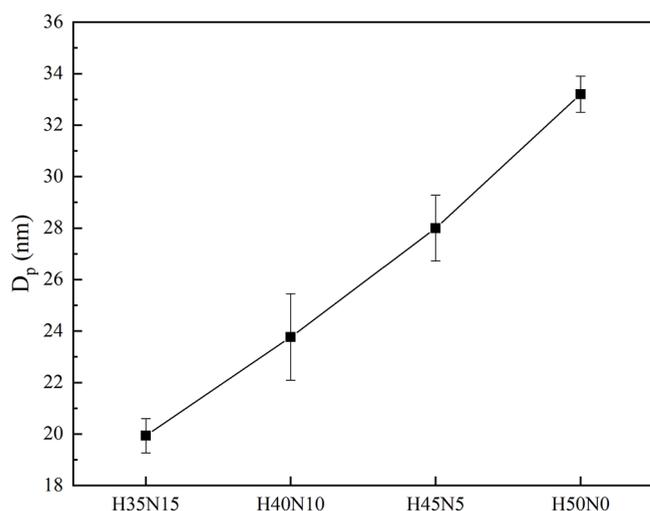


Figure 6. Average primary particle diameter of soot at different investigated flames; the error bars show the standard error.

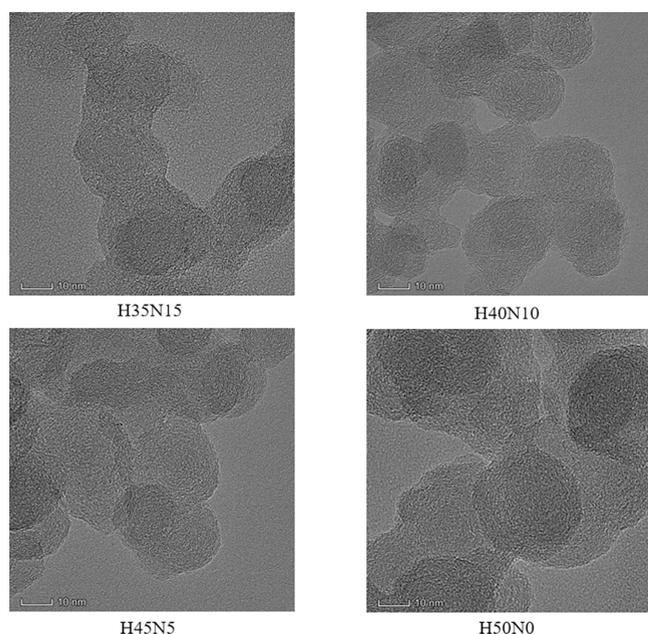


Figure 7. TEM images of soot morphology.

clarify the influences of H_2 addition on the concentrations of key species in the soot formation processes, chemical kinetic analysis was performed applying the diffusion flame reactor model of CANTERA software with the coupled H_2 - C_2H_2 -PAHs mechanism, and the calculation results were obtained through Python 3.8.2. The composition, inlet temperature and molar fraction of fuel and oxidizer are consistent with the acetylene laminar diffusion flame tested in this study.

The computed molar fraction profiles of A1 in laminar diffusion flames with the H_2 addition are shown in Figure 8. When X_{H_2} increases from 0 to 50%, the molar fraction of A1 increases. The formation of the first aromatic ring (A1) is the rate-determining step in the formation of PAHs, and A1 is also considered as an important precursor for soot formation.³⁹ The calculation results indicate that hydrogen could accelerate the soot formation by increasing the molar fraction of A1.

As shown in Figure 9, the molar fraction of the H radical increases monotonously with the H_2 addition. When X_{H_2} is

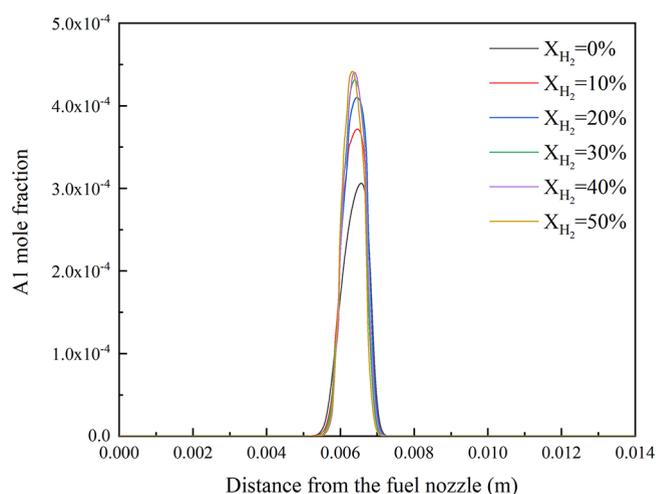


Figure 8. Computed molar fraction profiles of A1 in diffusion flames with H_2 addition.

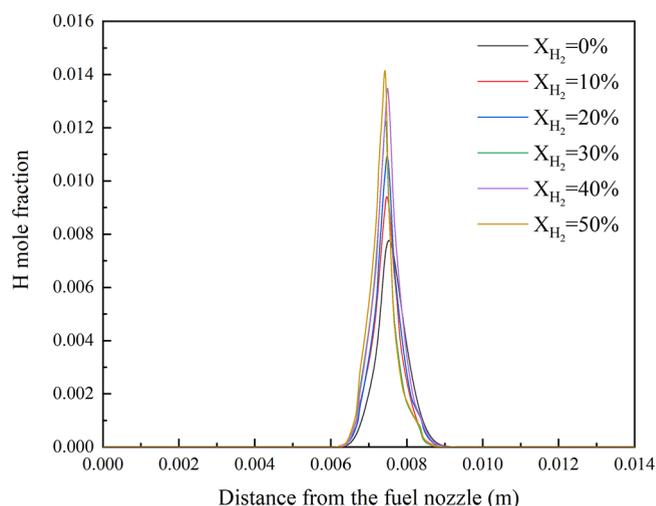


Figure 9. Computed molar fraction profiles of H in diffusion flames with H_2 addition.

increased from 0 to 50%, the concentration of A1 is increased by 43.4%. According to Frenklach,³³ the growth model of polycyclic aromatic hydrocarbons can be simplified as $A_i + H \leftrightarrow A_i^- + H_2$, $A_i^- + C_2H_2 \leftrightarrow A_iC_2H_2$, and $A_iC_2H_2 + C_2H_2 \rightarrow A_{i+1} + H$, which are the main reactions of the HACA path. The reaction $A_i + H \leftrightarrow A_i^- + H_2$ is an important way for PAHs to grow into soot, more H radicals could promote the reaction to generate more A_i^- , thus making PAHs grow continuously. Therefore, the concentration of A1 increases could correspond to the mass of soot collected increases in Figure 5. The results indicate that with the hydrogen addition, the increase of the molar fraction of A1 and H radicals promotes the PAH growth and soot formation.

Figures 10 and 11 show the computed molar fraction profiles of CO, CO₂ and H₂O with H_2 addition, respectively. As shown in Figures 10 and 11, the calculated molar fractions of CO and CO₂ decrease significantly with the hydrogen addition. The reduction of CO and CO₂ molar fractions indicate that H_2 addition does not promote the soot oxidation.³⁰ In addition, Figure 12 shows that the molar fraction of H₂O increases rapidly with the H_2 addition, which is because H₂O is the product of H_2 combustion. The presence of a large amount of

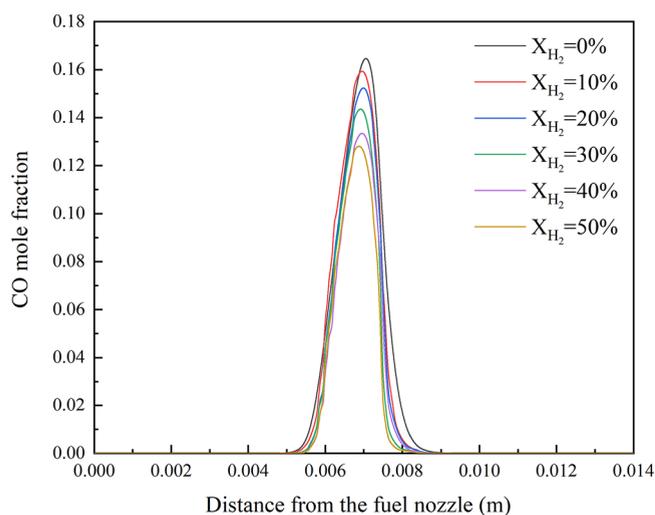


Figure 10. Computed molar fraction profiles of CO in diffusion flames with H₂ addition.

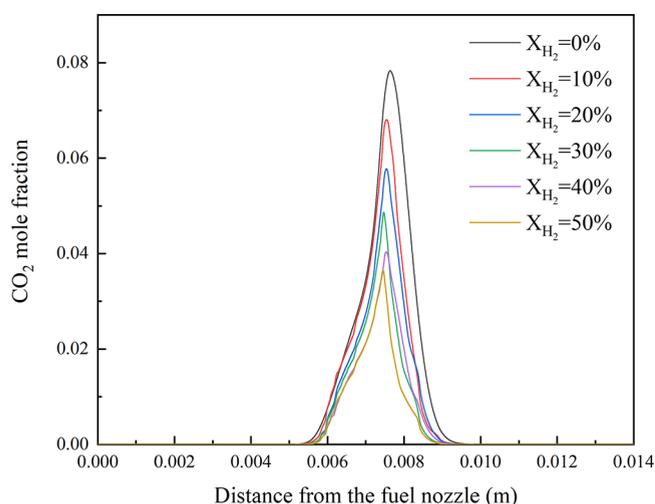


Figure 11. Computed molar fraction profiles of CO₂ in diffusion flames with H₂ addition.

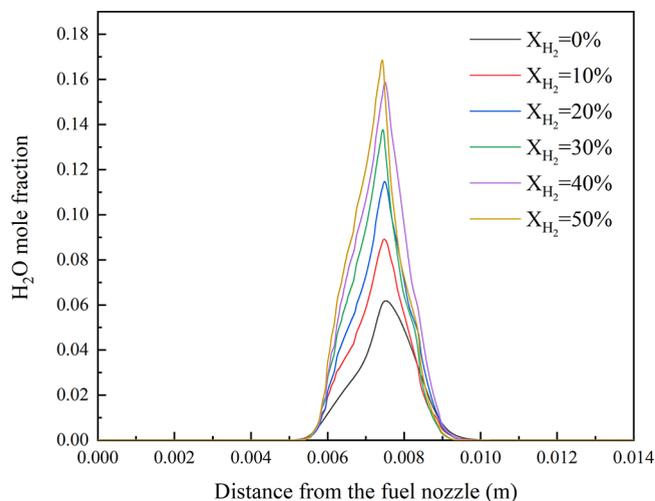


Figure 12. Computed molar fraction profiles of H₂O in diffusion flames with H₂ addition.

H₂O indicates that hydrogen has higher reactivity, which may lead to incomplete combustion of acetylene and increase the soot formation.

4. CONCLUSIONS

The effects of H₂ (between 0 and 50% by molar fraction while the remainder is nitrogen, N₂) addition on soot formation in acetylene (50% by molar fraction) laminar diffusion flames were experimentally and numerically investigated. The temperature profiles along the flame centerline, soot mass, and flame appearance were measured at each operating mode. A detailed H₂-C₂H₂-PAHs mechanism was also constructed to explore the influence of H₂ addition on the concentrations of key species. Overall, the results indicate that the addition hydrogen could promote the soot formation and emission. The result is different from previous studies and is the first report. Specifically, the following conclusion can be drawn:

1. The flame height increases, and the flame temperature decreases with the increase of H₂. The lower flame temperature could weaken the oxidation process of soot in the flame, thus leading to an increase in soot emission. The results indicate that H₂ addition could promote soot formation and emission in acetylene laminar diffusion flame.
2. When X_{H₂} = 30 to 50%, the mass of soot collected through the sampling hood increases significantly. Meanwhile, soot wings appear, with the soot breaking through the flame, the flame has obvious soot emission.
3. TEM results show that the size of soot particles increases and more aggregation of the soot particles are observed with the H₂ addition. The results indicate that hydrogen could increase the soot emission by promoting soot particle surface growth and particle aggregation.
4. The chemical kinetic analysis shows that the concentration of Al, H, and H₂O in C₂H₂ laminar diffusion flame increases with the addition of H₂. According to the HACA reaction, the increase of the molar fraction of Al and H radicals could promote the PAH growth and soot formation.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.3c01216>.

Chemical kinetic mechanism in CANTERA format (PDF)

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This research was supported by Zhejiang Provincial Natural Science Foundation of China under Grant No. LZ23E060001, LQ21E060003, and LZ21E060001.

NOMENCLATURE

T = temperature
 m_s = soot mass
 \dot{m}_s = mass per total sampling time
 D_p = average primary particle diameter
 X_{H_2} = molar fraction of hydrogen
 $F_{C_2H_2}$ = C_2H_2 flow rate
 F_{H_2} = H_2 flow rate
 F_{N_2} = N_2 flow rate
HAB = flame height above burner

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