Study on Soot Emission Characteristics of Methane/Oxygen Inverse Diffusion Flame

Runmin Wu, Fei Xie, Juntao Wei, Xudong Song,* Huijun Yang, Peng Lv, and Guangsuo Yu*

ABSTRACT: Inverse diffusion flame (IDF) is an effective and widely used reaction form in the process of noncatalytic partial oxidation (NC-POX) of gaseous hydrocarbons (such as natural gas and coke oven gas). However, soot is generated in the combustion chamber in the case of unreasonable feeding conditions, and thus causes serious damage to the wall and nozzle. In this study, the effects of the equivalence ratio ([O/C]e), the oxygen flow rate, and the Reynolds number on the soot and CH* emission characteristics of CH4/O2 inverse diffusion flame were comprehensively analyzed based on a hyperspectral imaging system. In addition, the relationship between CH* and soot is explored using Ansys Fluent simulation. The experimental results show that the soot radiation core generation area is located in the outer ring of the flame, and the radial distribution of the radiation intensity is bimodal. With the increase in [O/C]e, the initial position for soot radiation and the overall radiation intensity of soot decrease. In addition, the CH* radiation intensity decreases as [O/C]e increases, and CH* exists in the whole flame. The simulation results clearly show that the existence of CH* is conducive to soot production. The emission intensity and the core area of soot formation increase with the increase in the oxygen velocity. Additionally, the soot emission region increases and the flame tip changes from a round blunt to symmetrical tip with the increase in the Reynolds number.

1. INTRODUCTION

Natural gas is an attractive raw material for energy and chemical industries due to its environmental friendly advantage. Noncatalytic conversion of natural gas has been widely used for syngas production for its low feedstock consumption, a high calorific value, and no need for catalysts. In this process, the converter is the core apparatus for the synthesis gas reaction, and the operation parameters, environmental conditions, and combustion modes of the device are the most important influencing factors for reaction and ignition. A series of complex physical and chemical processes from a gas phase to a soot particle phase occur in the case of insufficient oxygen feeding and unreasonable conversion conditions as the natural gas conversion reaction proceeds. A large amount of soot radiation heat release is an important source of the reaction efficiency loss, and the soot attached to the furnace wall damages its structure after high-temperature ablation. Therefore, it is essential to study the radiation characteristics of soot in the conversion furnace, aimed at reducing the soot production in the reaction device.

The coaxial jet diffusion flame could be divided into normal diffusion flame (NDF) and inverse diffusion flame (IDF) depending on the gas inlet method. The central channel of NDF is filled with fuel, and the annular gap is filled with the oxidant. However, there are many limitations in investigating soot in the NDF. For example, the annular oxygen enwraps the soot generated in the central channel and further reacts quickly. On the contrary, the central channel of IDF is filled with the oxidant and the annular gap is filled with fuel. IDF is...
widely used in industrial furnaces and gas turbines because of its strong stability. The soot generated in IDF moves toward the lower-temperature region on the fuel side of the ring gap without being rapidly oxidized. Thus, this reaction method facilitates a systematic study of soot radiation. Moreover, the real-time monitoring of soot production can be realized through field optical measurement.

Optical diagnostics based on laser radiation, with higher temporal and spatial resolution, has become a widely used mainstream method for the diagnosis of soot in the flame. The measurement principle is that the soot particles in the flame are excited by the laser beam to release the spectral signal to describe the soot distribution characteristics and operation parameters. Therefore, a series of studies have been carried out using optical diagnostic techniques to explore the soot formation characteristics of the inverse diffusion flame. Xin et al. used a laser-induced incandescent (LII) method to measure the volume fraction of two-dimensional spatial resolution soot in turbulent flames of methane and ethylene and studied the distribution of soot on the vertical and horizontal planes. Bouvier et al. studied the formation and oxidation of soot in swirl-stratified premixed ethylene/air flame using LII and plane laser-induced fluorescence (PLIF) technology. The relationship between the initial, growth, aggregation, and oxidation processes of soot is described. De Iuliis compared the measured values of UV-excited LII and PLIF at four different detection wavelengths (340, 400, 450, and 550 nm) with the numerical simulation results in laminar ethylene–air coflow nonpremixed flame and analyzed the relationship between soot generation, soot radiation, and local temperature. The results showed that the numerical simulation results are in good agreement with the experimental results. Rodriguez proposed a new method to estimate the volume fraction field of soot in a laminar axisymmetric diffusion flame using diffuse line-of-sight attenuation (LOSA) that reconstructs the soot field through the data image captured by a camera.

However, the abovementioned detection methods require additional equipment such as a laser, an external light source, and an optical lens group, which are relatively complex. Therefore, another kind of flame-based emission spectrum technology is adopted. This method uses the flame spontaneous emission spectrum to detect hydrocarbon flame temperature, soot concentration, and radiation distribution characteristics, and its simplicity and efficiency have attracted increasing attention. The near-blackbody radiation generated by soot particles in the flame is located in the continuous radiation spectrum of visible and near-infrared bands. In view of this method, many scholars have studied the optical radiation characteristics of soot. Ayrancı used flame emission spectroscopy to determine the temperature, volume fraction, and the refractive index of soot and developed the existing soot diagnosis method based on flame emission spectroscopy. The temperature and volume fraction fields of soot were characterized in situ by one-dimensional chromatography reconstruction of soot emission spectroscopy (SSE). Liu analyzed the effect of water vapor addition on soot formation and flame performance, and it was found that water vapor not only affected soot formation and flame performance through dilution and a thermal effect but also affected flame performance through a chemical effect, which will inhibit soot formation. De Iuliis used multiwavelength emission technology to measure soot volume fraction and temperature in ethylene diffusion flame, and the results showed that the uncertainty derived from measuring the refractive index of soot is reduced using this technology. Sunderland studied the spectral radiation characteristics of soot in an ethylene/air turbulent diffusion flame, and the results show that the effect of soot continuous radiation on turbulent radiation is more significant than that of carbon dioxide. Modest verified the laminar flame and the turbulent flame by gas chemistry and a soot model and found that the measured temperature, soot volume fraction, and radiation heat flux were consistent.

In summary, the accurate measurement of radiation characteristics of soot particles plays an indispensable role in heat transfer analysis and optical diagnosis. Currently, although there is a certain understanding of the role of soot generation, and the researchers focus on study the variation of soot content and flame temperature in hydrocarbon fuel reactions with different parameters, whereas, the generation of soot
particles increases the radiative heat loss and decreases the reaction efficiency, etc. Therefore, the soot radiation during combustion has not been considered deeply and needs further study at present. Therefore, to visualize the soot formation characteristics in the combustion process of methane fuel more intuitively, this paper mainly studies the effects of \([\text{O/C}]_e\), the oxygen flow rate, and the Reynolds number on the structure of IDF and soot radiation intensity and analyzes the characteristics of soot formation. Combined with the GRI-Mech 3.0 mechanism, a two-dimensional numerical simulation of soot is conducted to verify that the radiation model can better predict the distribution of soot in the flame. This work will provide the basis for optimizing the reaction conditions and shows reference significance for heat transfer analysis in a converter furnace.

2. EXPERIMENTAL METHODS

2.1. Experimental Installation. Figure 1 represents the schematic diagram of a spectroscopy diagnostic system for an inverse diffusion soot laminar flame, which mainly consists of a flame generation system and a soot diagnosis system (SLR camera and a hyperspectral imaging system). The flame combustion system adopts the inverse diffusion flame (IDF) structure, which is a special type of diffusion flame. The combustion nozzle adopts a coaxial three-channel nozzle. The oxygen is fed into the central channel with an internal diameter of 2 mm. The annular interval is a methane channel with an inner diameter of 2.6 mm and an outer diameter of 3.6 mm. To prevent the interference of surrounding air in the research results, argon as the protective gas is introduced in the third channel for isolation. The relative position of the inverse diffusion flame burner and camera is shown in Figure 2. During the study, a mass flowmeter (CS200A, Beijing Qixing Flow Co, Ltd, China) is used to control the flow. The diagnostic system consists of an SLR camera (Sony A7M3) and a hyperspectral imaging system. The flame morphology and distribution are recorded by a camera, and the resolution is 693 × 425. A hyperspectral camera system includes a linear EMCCD hyperspectral camera (five-bell optics), an electronically controlled displacement table, and related software. Its principle is to use CCD image acquisition technology and beam splitting technology to scan the light source in the direction perpendicular to the slit and obtain the two-dimensional distribution of soot radiation of the complete image in the band. The spectral detection range of the hyperspectral camera is 400–1000 nm, the image resolution is 1004 × 1002, the spectral resolution is 2.73 nm, and the image pixel size is 0.1506 mm/pixel.

2.2. Experimental Conditions. The flame research conditions for different \([\text{O/C}]_e\), oxygen flow rates and Reynolds numbers \((Re)\) are shown in Table 1 and Table 2, respectively. In the study, the oxygen velocities selected were 3.58, 5.97, and 8.36 m/s to study the effects of different

| Table 1. Experimental Conditions of Soot at Different \([\text{O/C}]_e\) and Oxygen Velocities |
|---------------------------------------------------------------|
| case | \(\text{VO}_2\) (L/min) | \(\mu\text{O}_2\) (m/s) | \(\text{VCH}_4\) (L/min) | \(\mu\text{CH}_4\) (m/s) | \([\text{O/C}]_e\) | Re |
|------|-----------------|-----------------|-----------------|-----------------|-----------------|-----|
| 1    | 0.30            | 3.58            | 0.36            | 1.74            | 0.42            | 368 |
| 2    | 0.33            | 1.59            | 0.50            | 1.46            | 0.50            | 358 |
| 3    | 0.30            | 1.46            | 0.62            | 1.18            | 0.50            | 351 |
| 4    | 0.24            | 1.18            | 0.62            | 1.18            | 0.50            | 337 |
| 5    | 0.21            | 1.05            | 0.70            | 1.05            | 0.50            | 331 |
| 6    | 0.19            | 0.94            | 0.78            | 0.94            | 0.50            | 327 |
| 1    | 0.50            | 5.97            | 0.60            | 2.91            | 0.42            | 613 |
| 2    | 0.54            | 2.65            | 0.46            | 2.65            | 0.46            | 598 |
| 3    | 0.50            | 2.44            | 0.50            | 2.44            | 0.50            | 586 |
| 4    | 0.38            | 1.97            | 0.62            | 1.97            | 0.62            | 563 |
| 5    | 0.34            | 1.74            | 0.70            | 1.74            | 0.70            | 552 |
| 6    | 0.30            | 1.56            | 0.78            | 1.56            | 0.78            | 544 |
| 1    | 0.7             | 8.36            | 0.83            | 4.07            | 0.42            | 858 |
| 2    | 0.76            | 3.71            | 0.46            | 3.71            | 0.46            | 838 |
| 3    | 0.70            | 3.42            | 0.50            | 3.42            | 0.50            | 821 |
| 4    | 0.56            | 2.76            | 0.62            | 2.76            | 0.62            | 787 |
| 5    | 0.50            | 2.44            | 0.70            | 2.44            | 0.70            | 773 |
| 6    | 0.45            | 2.19            | 0.78            | 2.19            | 0.78            | 762 |

| Table 2. Experimental Conditions of Soot Measured at Different \(Re\) |
|---------------------------------------------------------------|
| case | \(\text{VO}_2\) (L/min) | \(\mu\text{O}_2\) (m/s) | \(\text{VCH}_4\) (L/min) | \(\mu\text{CH}_4\) (m/s) | \([\text{O/C}]_e\) | Re |
|------|-----------------|-----------------|-----------------|-----------------|-----------------|-----|
| 1    | 0.50            | 2.65            | 0.63            | 2.14            | 0.40            | 1428|
| 2    | 0.60            | 3.18            | 0.75            | 2.57            | 0.40            | 1714|
| 3    | 0.70            | 3.72            | 0.88            | 2.99            | 0.40            | 2000|
| 4    | 0.80            | 4.25            | 1.00            | 3.42            | 0.40            | 2286|
| 5    | 0.9             | 4.78            | 1.13            | 3.85            | 0.40            | 2571|
| 6    | 1.00            | 5.31            | 1.25            | 4.28            | 0.40            | 2857|
| 7    | 1.10            | 5.84            | 1.38            | 4.71            | 0.40            | 3143|
| 8    | 1.20            | 6.37            | 1.50            | 5.13            | 0.40            | 3428|

Figure 2. Structure of the inverse diffusion flame burner.
working conditions on the flame morphology and soot spectral radiation characteristics.

The calculation methods of \([O/C]_e\) and \(Re\) are calculated as follows

\[
[O/C]_e = \frac{n_{O_2}/n_{CH_4}}{(n_{O_2}/n_{CH_4})_e}
\]  

(1)

In the formula, \(n_{O_2}/n_{CH_4}\) is the gas mole ratio calculated by the actual flow rate of oxygen and methane and \((n_{O_2}/n_{CH_4})_e\) is the stoichiometric ratio when methane is completely consumed.

\[
Re = \frac{D_e u \bar{\rho}}{\bar{u}}
\]  

(2)

Each height value is normalized with equivalent diameters of the corresponding burner, which is calculated by formula \(3^{23,24}\)

\[
D_e = \frac{2(m_c + m_a)}{[a\rho(G_c + G_a)]^{1/2}}
\]  

(3)

where, \(D_e\) is the equivalent diameter of the nozzle, \(G_c\) and \(G_a\) are the momentum flux, \(m_c\) and \(m_a\) are the mass flows of the central and annular fluids, \(\bar{\rho}\) is the average density of fuel and the oxidant, and \(\bar{u}\) is the average velocity of methane and oxygen.

2.3. Image Processing. The distribution area of soot radiation intensity is the region covered with yellow when the flame occurs. Image J processing based on the color threshold at 100–220 is used to process the initial photos taken by a camera to get the soot distribution area, as shown in Figure 3.

![Figure 3. Image processing steps.](image-url)

The key steps of processing are as follows. First, the flame image is sharpened to make the flame profile clear and highlight the flame edge information.\(^{25}\) Then, the sharpened original image is converted into an 8 bit grayscale image without color information, and the brightness and contrast are adjusted to increase the recognition of the soot radiation area from the background radiation. Subsequently, the flame image is divided into a background region and a combustion region by threshold segmentation based on grayscale entropy and image sharpening. The black-and-white binary images of yellow soot radiation intensity distribution and the white background are obtained by threshold segmentation.\(^{26}\) The calculated pixel size is 3.60 mm/pixel; eventually, the area of the soot radiation region is extracted in the flame.

2.4. Numerical Model and Validation. To simulate the mechanism of soot generation in an inverse diffusion flame, the computational fluid dynamics (CFD) software Ansys is used to create a two-dimensional simulation geometry, the conceptual model of which is shown in Figure 4. The detailed chemical kinetic mechanisms obtained the GRI-Mech 3.0 for 37 species and 227 detailed chemical kinetic reactions after simplification to remove nitrogenous material.\(^{4}\) A three-channel axisymmetric nozzle consistent with the experiments is used to simulate the inverse diffusion flame, and a 1/2-section computational domain is constructed with axial and radial lengths of 100 ± 20 and 25 ± 20 mm, respectively. The diameter of the central oxygen channel is 2 mm, and that of the annular methane channel is 0.5 mm. The reaction is carried out in an argon flow to isolate the environment. The boundary conditions are set to the velocity inlet and the pressure outlet. The grid is encrypted in the nozzle outlet and the area near the axial symmetry. The number of grids is 20 879, 33 674, and 38 932, respectively, and it is verified that increasing the grid number has no significant effect on the numerical results. Therefore, 20 879 is chosen as the simulation object.

The nonpremixed model is used to simulate the combustion to obtain the convergence value and the standard \(k–\varepsilon\) model with the standard wall function is applied for viscosity calculations. The pressure–velocity coupling adopts the SIMPLE algorithm.\(^{27}\) The discrete transfer radiation (DTRM) radiation model is used to solve the radiation heat transfer equation. The two-step model in Ansys is chosen for the soot model, whose methane gas-phase chemical reactions, thermal, and transport properties predict the formation of nuclear particles, along with the formation of soot on the particle surface.\(^{28}\) The control equation is discretized by the control volume method.

To validate the numerical model, the soot distribution at different methane velocities is carried out to test the applicability of the model in the inverse diffusion flame. Figure 5 presents the two-dimensional distribution of soot radiation intensity versus the molar fraction, and it is clear from the results that the simulation is in general agreement with the experimental results. In this flame, the soot peaks are found in

![Figure 5. Comparison between the soot distribution by (a) experiment and (b) simulation under \(u_{O_2} = 5.97 m/s, Re = 598\).](image-url)
the midstream flanking region of the flame, which is similar to the simulated results of Zhang et al. 29 However, the experimental result reveals a small amount of soot generation at the end of the nozzle. In contrast, the simulated results show soot generation only at 5 mm from the end. The reason for this difference is that fuel reacts with air in an open environment.

3. RESULTS AND DISCUSSION

3.1. Visible Flame Image. Figure 6 reveals the flame morphology under different [O/C]e conditions when \( u_{O_2} = 5.97 \text{ m/s} \). The flame is composed of blue and yellow regions. With the increase in [O/C]e, the blue region increases and the yellow region decreases. Studies have shown that fuel is mainly pyrolyzed to generate soot radiation at high temperature in the yellow region, while in the blue region, it is mainly composed of free radicals. 30,31 When \([O/C]_e = 0.42\), the flame color changes from blue to yellow, and finally disappears along the direction of flame propagation. The oxygen content in the blue region of the flame root is sufficient, and the fuel is completely mixed with oxygen. The dark yellow region indicates that part of the soot begins to generate with the nucleation reaction of soot precursors, which is called the initial region of soot. As the flame height increases to the bright yellow region, the pyrolysis reaction is intensified due to the gradual increase in flame temperature. A large number of soot particles are finally formed by continuous condensation, growth, collision, and polymerization of soot precursors. This range is known as the zone of full of soot development. At the end of the flame, the yellow light gradually disappears. In this region, the oxidation reaction between soot and a OH* free radical in the flame occurs, and the oxidation rate of soot is greater than the formation rate, so the yellow light starts to weaken.

Figure 7 shows the continuous emission intensity information at different positions along the flame propagation direction captured by the hyperspectral camera. There are continuous emission and characteristic emission from specific particles in the wavelength range of 400–700 nm. The characteristic emission of CH* is 430 nm, while C2* emission shows characteristic peaks at 480, 523, and 566 nm. It can be noted from the figure that a and b are composed of free radicals such as CH* and C2*, and the intensity of free radicals generated at the root of flame a is high. Along the flame propagation to c, blue and yellow flame transition zones, the chemiluminescence of free radicals disappears along the flame propagation direction, forming a continuous spectral intensity that increases with the increase in the wavelength. In the yellow light symmetry region of the flame at e and f, the continuous emission intensity shows variations about the same. In addition, there is no free-radical characteristic emission peak in the visible range of 550–700 nm, and the blackbody intensity changes significantly. Therefore, to represent the
change of flame soot emission intensity, the intensity at a 620 nm wavelength is selected for study.

3.2. CH\* Intensity Distribution. In the lean oxygen reaction state ([O/C]_e < 1), the background emission is superimposed on the CH\* free-radical emission peak at 430 nm, including blackbody and CO2\* emission, and the emission accumulation near the CH\* band is significant. To eliminate the interference caused by background emission, the background emission at 430 nm is calculated by linear interpolation based on the background emission deduction method proposed by Karnani and Zhang et al. As shown in eqs 4 and 5, the calculation method of simultaneously deducting the blackbody and CO2\* emission can be obtained, as shown in eq 6.

\[
I_{\text{correction}} = I_{\text{measurement}} - (I_{\text{CO2}\*} + I_{\text{blackbody}}) \tag{4}
\]

\[
I_{\text{CH}^*} = I_{430\text{nm}} - (I_{\text{blackbody}} + I_{\text{CO2}\*}) \tag{5}
\]

where \(\lambda\) is the wavelength and \(I\) is the emission intensity at that wavelength.

\[
I_{\text{CH}^*} = I_{430\text{nm}} - (0.6361I_{420\text{nm}} + 0.3486I_{440\text{nm}}) \tag{6}
\]

The error between CO2\* emission intensity calculated by eqs 4 and 5 and the true value is less than 2%. Thus, the calculation method of simultaneously deducting the blackbody and CO2\* emission can be obtained, as shown in eq 6.

The CH\* emission intensity distribution of 430 nm after deducting the Prestock law is shown in Figure 8. Figure 8a shows that with the increase in [O/C]_e, CH\* emission intensity distribution decreases, and the axial and radial distribution also decreases. There is CH\* free radical at all flame fronts, so it can characterize the flame front. Figure 8b shows that the CH\* emission peak decreases to become stable when [O/C]_e is 0.60. The main formation pathways are: C2H + O2 → CO2 + CH\* and C2H + O → CO + CH\*. In the blue region upstream of the flame, the increase in the methane flow in the annular channel increases the CH\* production, and the CH\* production at the flame front is caused by the interaction between oxygen and C2H in the air. It indicates that the increase in hydrocarbon free radicals such as CH2 and C2H promotes the formation of CH\*, thereby enhancing the emission intensity of CH\*. When [O/C]_e > 0.62, it is known from the isoline that the CH\* generation area is stable, indicating that the formation of CH\* free radicals in the oxygen-rich state is less affected by [O/C]_e and less affected by the background emission.

3.3. Distribution of Soot Emission Intensity. Figure 9 is a two-dimensional image of soot emission of methane laminar inverse diffusion flame. With the increase in [O/C]_e, the emission area of soot decreases gradually and the emission intensity decreases. The peak intensity of soot mainly occurs in the outer annular region of the flame and is distributed symmetrically. This is mainly due to the fact that soot and polycyclic aromatic hydrocarbons formed by pyrolysis of outer fuels are not fully oxidized through the high-temperature zone inside the flame. When [O/C]_e ≤ 0.50, with the increase in [O/C]_e, the emission height of soot decreases rapidly under the same oxygen flow, while the radial diffusion is basically unchanged. When [O/C]_e > 0.50, the methane flow continues to decrease, the flame height remains stable, and the emission height of soot decreases.
distribution of soot emission intensity and the content area disappear. This is caused by the increase in $[O/C]_o$, which makes the oxygen content to increase relatively and a complete reaction of the fuel to form $CO_2$ and $H_2O$, with only a small amount of fuel reacting at high temperatures to form soot.

It can be seen that the CH* intensity mainly occurs in the blue region upstream of the flame, and the CH* intensity
distribution is basically consistent with the soot intensity distribution. Its main reason is that when \([O/C]_\text{e}\) is low, the carbon content in the fuel increases, and a large number of \(\text{CH}^*\) free radicals are generated, which then gradually agglomerate and grow into soot precursors at high temperature, thus forming soot particles. With the increase in \([O/C]_\text{e}\), the flow rate of methane decreases, which makes the \(\text{CH}^*\) free-radical content to gradually decrease.

Figure 10 shows the radial distribution of soot emission intensity at different heights from the nozzle. As \([O/C]_\text{e}\) increases, the soot intensity decreases. The soot emission is clearly bimodal along the radial position of the flame, with the peak intensity occurring in the symmetrical area of the outer flame. As the flame height increases to a position close to the flame tip, the soot intensity increases and then decreases, and the bimodal distribution gradually decreases until it shows a single peak distribution. The soot emission intensity is less than 2000 mW Sr\(^{-1}\) m\(^{-2}\) when the flame height is lower than 2.2\(D_\text{e}\), indicating that the emission signal of soot particles in the flame is greatly weakened. This location is defined as the nucleation region for PAHs, and a large number of soot particles with near-blackbody properties are not formed. In the middle and upper part of the flame, PAHs begins to evolve to soot particles, resulting in a large number of soot particles in the outer ring region of the flame reaching a peak in the middle part of the flame. Then, as the height increases, the soot particles are oxidized and the emission intensity weakens gradually. Olofsson\(^{36}\) found that soot particles below 10 mm were mainly dispersed small particles, and more aggregates appeared at positions higher than 10 mm, which was relatively consistent with this study.

The intermediate substances of soot model nucleation precursors in Ansys simulations are mainly \(\text{C}_2\text{H}_2\) and \(\text{C}_2\text{H}_4\), whose production paths are \(\text{CH} + \text{CH}_4 \rightarrow \text{H}^+ + \text{C}_2\text{H}_4\) and \(\text{CH} + \text{CH}_2 \rightarrow \text{H}^+ + \text{C}_2\text{H}_2\). It can be noted from Figure 11 that with the ongoing reaction, the formation of \(\text{C}_2\text{H}_2\) and \(\text{C}_2\text{H}_4\) is mainly located upstream of the flame, and the distribution of soot nucleus particles is located in the region of 12–22 mm. The initial formation of soot particles starts at 10 mm away from the nozzle end, which is basically consistent with the experiment. Soot nucleation is the core procedure of the whole soot particle formation. In the present simulations, the first polycyclic aromatic hydrocarbon, benzene ring (A\(_1\)), is chosen as the soot precursor. Dworkin\(^{37}\) concluded that the main pathway of A\(_1\) formation is

\[
\text{C}_2\text{H}_4 \rightarrow \text{C}_2\text{H}_2 \rightarrow \text{CH}_3 \rightarrow \text{C}_2\text{H}_3 \rightarrow \text{A}_1 \rightarrow \text{PAHs}
\] (7)

The formation of PAHs by hydrocarbon-abstraction-C\(_2\)H\(_2\)-addition (HACA) kinetic mechanism is
$A_1(+C_2H_2) \rightarrow A_1C_2H(\pm C_2H_2) \rightarrow A_2 \rightarrow \text{PAHs}$  \hspace{1cm} (8)

It is concluded that the nucleation and surface growth of soot are mainly generated by $C_nH_2$ and $C_nH_4$, which play a key role in the formation of soot precursors.

Figure 12 presents the simulated two-dimensional distribution of a CH* free radical and soot. It has long been confirmed that the small radicals, such as CH*, OH*, and $C_2H_4$, play a crucial role in the formation of PAHs and benzene isomers. One of the most reactive radical species is CH*, which is related to soot formation in hydrocarbon flames. In the gas phase, the CH* group is able to promote the growth of cyclic unsaturated hydrocarbon molecules and plays a major role in the first ring-forming step in the growth of PAHs, which enhances the tendency of soot formation, in turn. The chemical reaction schemes are summarized mainly by the CH-addition-H-elimination mechanism:

$CH + C_nH_m \rightarrow C_{n+1}H_{m+1} + H$. The abovementioned facts are characteristics of CH* radical reactions that can lead to the formation of benzene isomers.

3.4. Effect of the Oxygen Flow Rate on Soot.

The distribution of the peak intensity of soot emission with [O/C]e at different oxygen flow rates is shown in Figure 13. With the increase in [O/C]e, the peak value of soot intensity gradually decreases to 0. The peak intensity of soot increases with the increase in the oxygen flow rate under the same [O/C]e. With the increase in the oxygen flow rate, the reaction is intensified and the temperature increases. The soot generated by pyrolysis of the fuel at high temperatures increases the intensity of blackbody emission, enhancing the flame brightness. When $u_{O_2} = 3.58$ m/s, the peak value of emission intensity changes dramatically, and with the increase in [O/C]e, the peak value of soot emission is basically stable when [O/C]e is 0.62. This is because of rapid mixing of the fuel/oxidant reaction, resulting in the flame height that is relatively short.

Figure 14 shows the axial distribution of soot emission intensity under different oxygen flow rates, where the emission height is defined as the distance from yellow light to the end of yellow light at the flame tip. The figure shows that the change of oxygen–fuel ratio has a significant effect on soot emission height. Based on this, emission height can be expressed as

$$H_{soot}/D_e = \left(0.0975 + 1.05V_{O_2} - 1.25V_{O_2}^2\right) \times [O/C]_e^{-\left(6.975 - 7.5V_{O_2} + 7.5V_{O_2}^2\right)}$$  \hspace{1cm} (9)

With the increase in [O/C]e, the soot emission height decreases significantly under different oxygen flow rates. The reason for this phenomenon is that the lower methane flow makes the flame height gradually shorter, resulting in a shorter soot emission zone height. When [O/C]e > 0.60, $H_{soot}/D_e$ remains invariant, which is due to the decrease in the methane flow rate and the increase in the relative oxygen diffusion rate, so the mixing reaction rate of fuel and oxygen increases and the methane reaction is complete. With the increase in the oxygen flow rate, $H_{soot}/D_e$ decreases. When [O/C]e = 0.42, $H_{soot}/D_e$ increases from 24 to 34, with a growth rate of 41.7%. When the fuel flow rate is large, the oxygen diffusion rate is limited, so methane needs to diffuse to a higher position to react with oxygen. It demonstrates that the variation in the soot emission intensity height is caused by different oxidant flow rates, and the variation trend of the flame length with fuel is basically the same. Therefore, the increase in the soot emission height is the result of the combined effect of the relative flow rate of fuel and the flow rate of the oxidant.

Figure 15 shows the distribution of the soot emission intensity area in the flame, which reflects the spatial distribution of soot in the reaction process. The fitting curve can be expressed as eq 10

$$A = (-3.7 + 18.8SV_{O_2} - 18.12SV_{O_2}^2) \times [O/C]_e^{-\left(12.4 - 2.5V_{O_2} + 26.25V_{O_2}^2\right)}$$  \hspace{1cm} (10)

The region of emission intensity reduces gradually with the increase in [O/C]e, which is consistent with the changes in Figures 13 and 14 basically. Under different oxygen flow conditions, the change of the region of soot emission intensity is the most prominent when [O/C]e is adopted from 0.42 to 0.46. The distribution area of soot emission is the largest at $u_{O_2} = 8.36$ m/s in the flame. This indicates that a higher fuel flow causes a longer and a wider flame before the flow reaches turbulent conditions. As a result, larger areas of soot are created. In consequence, at a higher fuel flow, the flame surface increases. Based on the abovementioned conclusions, with the
increase in the oxygen flow, the emission height, the emission area, and the peak intensity of soot tend toward stability when $[O/C]_e \geq 0.6$.

### 3.5. Effect of the Reynolds Number on the Emission Intensity of Soot

Figure 16 shows the morphological changes of the inverse diffusion flame formed under different Reynolds numbers ($Re$) with an $[O/C]_e$ of 0.4. As $Re$ increases, the height of the inverse diffusion flame increases progressively. The complete reaction of oxygen and methane at the root of the flame makes the flame blue and the formation height of the blue flame increases with the increase of $Re$. At the front and downstream of the inverse diffusion flame, a large amount of soot is generated due to the lack of oxygen, and soot emission leads to a bright yellow region in the upper part of the flame, which has obvious soot characteristics. With the increase in $Re$, the diffused methane gas to the axial direction of the internal flame reacts with the pure oxygen in the inner channel. When $Re < 2000$, the flame is stable and the tip is circular. The flow state enters the transition zone as the $Re$ increases, the flame geometry turns yellow and bright, and more slender needle-like symmetrical small tips appear, which are slender and stable.

Figure 17 shows the distribution of soot emission in the axial direction of the inverse diffusion flame at different Reynolds numbers that increases and then decreases in the direction of axial flame propagation. The core region of soot formation is gradually away from the nozzle end and the height of this region increases gradually. A result of the increase in the oxygen velocity promotes the mixing of root fuel and oxygen. However, in the middle and lower reaches of the flame, the soot particles move with the fluid to increase the distribution range. When $Re < 2286$, there is no significant change in the peak emission intensity of soot along the axis, which shows that the flame is stable in a laminar flow, and $Re$ has little effect on soot formation. When the fluid is in transition, the peak intensity of soot increases with the increase in $Re$, and when $Re$ gradually increases, the emission intensity of soot is widely distributed in the axial direction. There is a small peak at 5 mm away from the nozzle exit, which is mainly caused by the formation of a recirculation zone at the end of the nozzle. The soot emission intensity is always maintained at 2000 W Sr$^{-1}$ m$^{-2}$.

The peak intensity of soot increases with the Reynolds number, as shown in Figure 18. The peak soot intensity increases monotonically with increasing fluid Reynolds number. When the gas phase transits from laminar flow to

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**Figure 15.** Variation of soot emission intensity area distribution.

**Figure 16.** Methane flame image varies with the Reynolds number.

**Figure 17.** Distribution of the peak emission intensity of soot.

**Figure 18.** Distribution of the axial soot emission intensity at different Reynolds numbers.
turbulence, the emission intensity has a slow buffer, and when it reaches the transition region, the change degree is obvious. Combined with Figure 17, it is shown that the flame volume increases with the increase in the flow velocity before the turbulent state. In addition, the soot emission has little dependence on $Re$.

4. CONCLUSIONS

In this work, the soot emission characteristics of methane—oxygen inverse diffusion flame were studied in detail using hyperspectral imaging technology. The main conclusions are summarized as follows:

1. The flame is composed of free-radical emission in the blue region and continuous soot emission in the yellow region. The CH* free radical is distributed on the flame front, and its intensity is consistent with the variation trend of soot intensity with $[O/C]$e. In addition, the interference of background emission on free-radical emission detection is greater in the case of low values of $[O/C]$e. The simulation results verify that CH* radicals play a vital role in the formation of soot; it can be inferred that the CH* free-radical content change is also applicable in macromolecular hydrocarbon fuels.

2. The emission height and the area of the soot in the flame increase with the increment in the oxygen velocity, and the distribution is inversely proportional. In laminar flames, the high oxygen flow rate leads to a longer and wider flame, which, in turn, results in a larger amount of soot being produced.

3. As the Reynolds number increases, the emission intensity of the soot increases and the distribution area in the flame gradually enlarged. As the gas fluid enters the turbulent transition zone, the soot emission intensity changes rapidly. It indicates that the flow state has a strong influence on the generation of soot.

AUTHOR INFORMATION

Corresponding Authors

Xudong Song — State Key Laboratory of High-efficiency Utilization of Coal and Green Chemical Engineering, College of Chemistry and Chemical Engineering, Ningxia University, Yinchuan 750021, China; orcid.org/0000-0002-8211-409X; Email: xdsong@nxu.edu.cn

Guangsu Yu — State Key Laboratory of High-efficiency Utilization of Coal and Green Chemical Engineering, College of Chemistry and Chemical Engineering, Ningxia University, Yinchuan 750021, China; Institute of Clean Coal Technology, East China University of Science and Technology, Shanghai 200237, China; orcid.org/0000-0003-4085-9736; Phone: +86-0951-2062008; Email: gsyu@nxu.edu.cn

Authors

Runmin Wu — State Key Laboratory of High-efficiency Utilization of Coal and Green Chemical Engineering, College of Chemistry and Chemical Engineering, Ningxia University, Yinchuan 750021, China; orcid.org/0000-0002-2343-6574

Fei Xie — State Key Laboratory of High-efficiency Utilization of Coal and Green Chemical Engineering, College of Chemistry and Chemical Engineering, Ningxia University, Yinchuan 750021, China

Juntao Wei — Joint International Research Laboratory of Biomass Energy and Materials, Co-Innovation Center of Efficient Processing and Utilization of Forest Resources, College of Materials Science and Engineering, Nanjing Forestry University, Nanjing 210037, China

Huijun Yang — Chinese Energy Ningxia Coal Industry Co., Ltd, Yinchuan 750021, China

Peng Lv — State Key Laboratory of High-efficiency Utilization of Coal and Green Chemical Engineering, College of Chemistry and Chemical Engineering, Ningxia University, Yinchuan 750021, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.1c02789

Notes

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