Experimental Study on Flame Propagation Characteristics of Premixed Laminar Combustion of Hydrogen-rich Coke Oven Gas

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Abstract—Coke oven gas (COG) is the main by-product of the coking industry, with a hydrogen content of more than 50%. Upon purification, COG can be used as a hydrogen-rich, efficient, high-quality alternative fuel, to improve the energy structure, alleviate global warming, and reduce polluting emissions. In the present paper, a constant-volume combustion experiment bench was established to carry out the premixed laminar combustion experiments of the COG-air mixture. A high-speed camera was used to record the flame propagation process and the effects of the influencing factors such as fuel-air equivalence ratio, initial pressure and \( H_2 \) concentration on the flame speed. The results indicated that when \( \Phi = 1.1 \), i.e., for a relatively concentrated fuel, the flame speeds of the stretched and unstretched flame both reached a maximum. With the increase in initial pressure, the flame speeds of both the stretched and unstretched flame decreased. As the \( H_2 \) concentration increased, the propagation speeds of the stretched and unstretched flame both increased and the enhancements were more obvious; mainly since hydrogen combuts faster than methane.

1. Introduction
China has the largest coke production in the world, accounting for around 60% of global coke production. The coke production generates a large amount of COG, which is also a remarkably rich energy source. The main components of COG are hydrogen (55% to 59%) and methane (24% to 26%), as well as a small amount of gases like carbon monoxide and nitrogen \(^1\), which represents a typical hydrogen-rich gas mixture. In 2019, China's coke output was 471 million tons; 427 cubic meters of COG as a by-product was produced per ton of coke \(^2\) and the annual COG production exceeded 200 billion cubic meters. About 30 billion cubic meters of COG are directly discharged and burned every year, causing a direct economic loss of around 10 billion yuan \(^3\). In September 2020, China proposed that CO\(_2\) emission would reach peak value before 2030 and would achieve carbon neutrality before 2060 \(^4\). If the COG generated by the coke production process is effectively recycled, it will be of great significance in realizing resource recycling, establishing low-carbon environments, and creating economic benefits.

At present, the use of COG has been mainly focused on hydrogen production, power generation, methanol production, ammonia synthesis, natural gas production and use as a fuel gas. Considering that the \( H_2 \) content of COG was relatively high, the use of COG to produce \( H_2 \) is an advantageous resource and has a lower price \(^5\). There are three main power generation modes that use COG, i.e., steam turbine, gas turbine and internal combustion engine, among which the internal combustion engine has the highest efficiency in power generation, being about 30% to 40% \(^6\). The catalytic
conversion of hydrocarbons in COG, into a certain proportion of CO and H₂, can produce methanol; generally, 2000 m³ ~ 2200 m³ COG can produce 1 ton of methanol [7]. COG produces hydrogen first, then ammonia with nitrogen gas is obtained under certain temperature and pressure and finally, urea is synthesized. Urea as fertilizer is an indispensable consumable in agricultural production [8-9]. The CH₄ purity of natural gas produced by COG is over 99% and the existing gas supply pipeline can be used. Therefore, it has the advantages of simple storage and easy transmission, which can be directly merged with the natural gas pipeline network of the West-East Gas Pipeline project to supply to cities [10-11].

Based on the physical and chemical parameters of COG, Bo et al.[12] established a model to describe the characteristics of COG for vehicles. The calculations indicated that COG had a higher calorific value and the engine could manifest a good overall performance when the COG was directly used as fuel. Ortiz Imedio R. et al. [13] carried out experimental studies on the engine of a Volkswagen Polo car using COG, pure H₂ and pure CH₄. The obtained data indicated that COG combined the advantages of pure H₂ and pure CH₄ and manifested remarkable engine performance and emission results, which were comparable to those of pure gas. Hongqing Feng et al. [14] analyzed the effectiveness of using COG and spark to ignite the engine. The results indicate that the delay of ignition timing and the increase in compression ratio can restrain the irreversibility of the combustion process of the engine using COG. Huang Kai et al. [15] experimentally studied the turbulent combustion process of stoichiometric mixing of H₂/CH₄/air in a spherical, constant volume combustion chamber. The relationship between the combustion velocity and relative combustion mass rate was discussed, and the effects of turbulence intensity and hydrogen volume fraction on the maximum combustion velocity were analyzed. Zhenxing Li et al. [16] applied O₂/CO₂ oxidants in the combustion chamber and found that they could significantly reduce the NOₓ emissions from the COG combustion.

In China, the research and development of the COG engine has begun and is being gradually implemented. Shandong province has manufactured several COG-generating sets, some areas in Yunnan province have constructed COG power stations, and the Guizhou province has been partially commercialized and has built a COG refueling station for 30 COG-driven buses and 100 COG-driven taxis.

So far, no in-depth experimental and theoretical studies have been carried out on the combustion of COG for internal combustion engines. Therefore, we carried out experimental studies on the flame propagation characteristics of premixed laminar combustion of COG in a constant-volume combustion bomb. The combustion and flame propagation mechanisms were investigated to obtain supporting data basis to design COG engines.

2. Experimental equipment

The constant-volume combustion equipment in the experiment consists of a constant-volume combustion bomb, a system to mix gas, an ignition system, a high-speed data acquisition system, a high-speed camera and a timing-control system. Among them, the constant-volume combustion bomb is the container where combustion occurs. The mixture preparation system can realize the mixture preparation of different equivalent ratio, various initial pressure and different hydrogen concentration. The high speed photography system and the high speed data acquisition system are used to record the flame propagation image and the combustion pressure change curve during the combustion process respectively. The timing-control system is used to coordinate a series of actions, including ignition, shooting by high-speed camera and recording by high-speed data acquisition system. Figure 1 shows the layout of the entire experimental setup.
3. Experimental methods
The study used a mixture of H₂ and CH₄ to simulate COG and used a mixture of O₂ (21%) and N₂ (79%) to simulate air. A premixed laminar combustion test of the COG-air mixture was carried out in a constant-volume combustion bomb. By varying the equivalence ratio \( \Phi \), initial pressure \( P_0 \) and H₂ concentration, the effects of these factors on the flame propagation characteristics of premixed laminar combustion were investigated. See Table 1 for details.

| Parameters of system | Values |
|----------------------|--------|
| Fuel-air equivalence ratio, \( \Phi \) | 0.8~1.2 |
| Initial pressure, \( P_0 \) | 300~500kPa |
| Initial temperature, \( T_0 \) | 298K |
| Coefficient of residual gas | 0 |
| Spark plug and electrode gap | USA, STITT, 0.65mm |
| Ignition position | the center of one ending surface of the constant-volume cylindrical bomb |
| COG 1 | H₂: 0%, CH₄:100% |
| COG 2 | H₂:58%, CH₄: 42% |
| COG 3 | H₂: 74%, CH₄: 26% |

4. Data processing
In the spherical diffused flame of COG, the flame speed, \( S_n \), is the derivative of the flame radius with respect to time, obtained by the following equation:

\[
S_n = \frac{R_{t+1} - R_{t-1}}{t_{t+1} - t_{t-1}}
\]

where, \( R \) denotes the flame radius shown in the photography and \( t \) denotes the time.

The stretched rate of flame, \( \alpha \), is defined by the following equation, as the derivative of the logarithmic value of an infinitely small area, \( A \), on the flame surface, with respect to time:

\[
\alpha = \frac{d(\ln A)}{dt} = \frac{dA}{Adt} = \frac{2}{r} \times \frac{dr}{dt} = \frac{2}{r} S_n
\]

In the initial stage of the diffusion of the COG flame, the flame front shows a relatively regular spherical shape. Herein, the stretched flame propagation velocity has a linear relationship with the flame’s stretched rate, given by:
\[ S_f - S_n = L_o \alpha \]  \hspace{1cm} (3)

where, \( S_f \) denotes the propagation speed of the unstretched flame; the result can be obtained when \( \alpha = 0 \); and \( L_o \) denotes the negative value of the Markstein length, representing the stability of flame.

5. Results and discussion

The test was carried out in a constant-volume combustion device. A high-speed camera was used to acquire the photos of flame propagation and then photoshop software was used to process these photos. The effects of equivalence ratio, initial pressure and H\(_2\) concentration on flame propagation characteristics were obtained, as follows:

Figure 2 show that the fuel was sufficient at the moment of ignition and the flame color was light blue. During the propagation process, the flame first turned red and then yellow. Finally, a large area of incandescent light appeared in the center, thereby forming a temperature gradient that gradually increased from the outer flame to the inner flame. When the flame turned dark red, it indicated that the fuel almost ran out. After the flame, there were gray smoke and tiny water droplets on the quartz glass.

![Fig. 2 Photos of premixed laminar combustion of COG 2-air mixture (P\(_0\)=300kPa, T\(_0\)=298K, \( \Phi = 0.9 \), shooting speed = 2000 fps)](image)

5.1. Effect of equivalence ratio on the propagation velocity of flame

The diameter of the constant-volume cylindrical bomb used in the study was 130 mm and the ignition position was at the center of the top of the constant-volume combustion bomb. The flame contacted the bomb wall as the flame radius was greater than 65 mm. To obtain an accurate behavior of the premixed laminar combustion, only the case where the flame radius was less than 65 mm was studied.
Figure 3 shows the relationship between the stretched flame propagation velocity and flame radius. The ignition induced a fluctuating effect on the flame development, which resulted in a larger error in the calculation of the initial ignition stage. Therefore, only the curves are shown only for flame radius in the range of 10–60 mm. For all equivalence ratios in the test, the stretched flame propagation velocity first increased and then decreased with the increase in the flame radius. Theoretically, the flame speed tends to decrease when the COG mixture is too concentrated or too diluted. The maximum stretched flame propagation velocity in the measured data appeared at $\Phi = 1.1$. The reason may be that the concentration of the activated centers in the flame was higher when the fuel was concentrated.

Figure 4 shows the relationship between the propagation speed of the unstretched flame and the equivalent ratio of the COG gas-air mixtures. The propagation speed of the unstretched COG 1 increased slightly with the increase in the equivalence ratio, but the enhancement was not obvious; the flame speed showed a downward trend after $\Phi > 1.1$. The propagation speed of unstretched COG 2 flame first increased, decreased and then increased again with the increase in the equivalence ratio.

The propagation speed of the unstretched COG 3 flame increased significantly with the increase in the equivalence ratio. For all the three COG-air mixtures, the maximum propagation speeds of the unstretched flame appeared at $\Phi = 1.1$, indicating that the flame speed was the highest when the mixture was relatively concentrated. The main reason was that the fuel itself was sufficient, so the combustion was more intense, which led to more heat released and higher combustion temperature.
5.2. Effect of initial pressure on the propagation velocity of flame

![Graph](image)

Fig. 5 The relationship between the propagation speed of stretched flame and the flame radius under various initial pressures

![Graph](image)

Fig. 6 The relationship between propagation speed of unstretched flame and initial pressure

Figure 5 indicate that as the flame radius increased, the stretched flame propagation velocity first increased and then decreased. As the pressure increases, the dissociation is reduced and the flame temperature increases, thus increasing the flame propagation speed. However, less dissociation suggests the decrease of active roots and the decrease of flame propagation velocity; thus, the two effects are mutually restrictive. From the experimental results, it can be seen that the active root plays a more significant role.

Figure 6 shows the relationship between the propagation speed of the unstretched flame and the initial pressure. The propagation speed of the unstretched flame is shown to decrease with the pressure and increase with the H₂ concentration.

5.3. Effect of H₂ concentration on the propagation velocity of flame

![Graph](image)

Fig. 7 Variation curve of stretched flame propagation speed with flame radius at various hydrogen concentration

![Graph](image)

Fig. 8 Unstretched flame propagation speed versus hydrogen concentration

Figure 7 shows the comparison of the propagation speed of stretched flame versus flame radius. The stretched flame propagation velocity of three COG-air mixtures all show a trend of first increasing and then decreasing and reach a peak at around 23 mm. Additionally, with the increase in H₂
concentration of the COG, the stretched flame propagation was faster, which was mainly determined by the combustion-supporting role of the hydrogen gas.

Figure 8 shows the relationship between the flame speed of the unstretched flame and H₂ concentration of the COG-air mixtures with different equivalence ratios at an initial temperature of 298 K and initial pressure of 300 kPa. With the increase in H₂ concentration, the speeds of the unstretched flame all showed an increasing linear trend. For COG 1, the propagation speed of the unstretched flame was not affected by the variation in the equivalent ratio, while COG 3 was largely affected by the equivalent ratio. The H₂ concentration had a more obvious effect on the propagation speed of the unstretched flame.

6. Conclusion

Based on the results and discussions presented above, the conclusions are obtained as below:

1) With the increase in the equivalence ratio, the propagation speeds of stretched and unstretched flame both increased accordingly, and the maximum values appeared at Φ = 1.1. As the flame radius increased, the stretched flame propagation velocity showed a trend of first increasing and then decreasing, reaching a maximum at about 23 mm and then gradually decreasing. When it approached the maximum flame radius, the propagation speed of the stretched flame decreased rapidly.

2) With the increase in the initial pressure, the propagation speed of the stretched and unstretched flame both decreased. As the flame radius increased, the propagation speed of stretched flame first increased and then decreased. The two speeds were closer at a radius of 55 mm.

3) As the H₂ concentration increased, the propagation speeds of stretched and unstretched flame both increased and the enhancements were more obvious, which mainly resulted from the combustion-supporting role of hydrogen gas.

4) In future studies, the functionality of the constant-volume combustion test bench can be further expanded, and a turbulence-generating device can be introduced to realize the premixed turbulent combustion.

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