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Numerical Simulation of the Influence of CO_2 on the Combustion Characteristics and NO_X of Biogas

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The existence of inert gases such as N_2 and CO_2 in biogas will reduce the proportion of combustible components in syngas and affect the combustion and NO_X formation characteristics. In this study, ANSYS CHEMKIN-PRO software combined with GRI-MECH 3.0 mechanism was used to numerically simulate the effects of different CO_2 concentrations (CO_2 volume ratio in biogas is 0–41.6%) on flame combustion temperature, flame propagation speed and nitrogen oxide formation of complex biogas with low calorific value. The results showed that when the combustion reaches the chemical equilibrium, the flame combustion temperature and flame propagation speed decrease with the increase of CO_2 concentration, and the flame propagation speed decreases even more slowly. Meanwhile, the molar fraction of NO at chemical equilibrium decreases with the increase of CO_2 concentration and the decrease is decreasing, which indicates that the effect of CO_2 concentration in biogas on NO is simpler. While the molar fraction of NO_2 does not change regularly with the change of CO_2 concentration, the effect of CO_2 concentration in biogas on NO_2 is complicated. The highest molar fraction of NO_2 was found at chemical equilibrium when the CO_2 concentration was 33.6%, when the target was a typical low calorific value biogas.

Keywords: simulation, biogas, CO_2 concentration, combustion characteristics, NOX

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INTRODUCTION

Biomass is a kind of clean and convenient energy with rich reserves, which is a promising green renewable energy (Zhang et al., 2005). Nowadays, direct combustion is a main utilization method for biomass (Yang et al., 2020). However, it has the disadvantages of low utilization ratio, massive content of dust and NO_X in flue gas (Zhao and Su, 2019; Wang et al., 2020). To solve above issues, the biogas obtained from pyrolysis or gasification of biomass can be combusted with pulverized coal as boiler fuel, which not only reduces the consumption of coal, but also decreases the NO_X emission significantly (Zhang et al., 2017). Therefore, biomass gasification is considered as a more effective, promising and valuable application mode for biomass utilization (Cao et al., 2019). However, the composition of biogas is complex, especially the existence of inert gases such as CO_2 and N_2 will have a great impact on the combustion characteristics of flame and the formation of NO_X (Chu et al., 2021).

In recent years, many scholars have studied the effects of simple component gases on flame characteristics and NO_X emission in laminar premixed combustion: Studies by many scholars have shown that the increase of H_2 content will lead to higher combustion temperature and laminar flame speed, but the emission of nitrogen oxides will also increase correspondingly (Azimov et al., 2011; Wang et al., 2021). The premixed combustion of syngas under different H_2/CO ratio was studied, the results suggested that increasing the H_2/CO ratio led to a decrease in the temperature as well as the NO concentration near the flame (Asgari et al., 2017). The combustion characteristics of NH_3/H_2/air, H_2/CO/syngas, propane/hydrogen/air and other mixtures have also been extensively studied (Tang et al., 2008; Nozari and Karabeyoglu, 2015; Chen and Jiang, 2021).

Although many scholars have studied the flame combustion characteristics of biogas, most of them are limited to study some kind of combustible gas, or changing the ratio of two kinds of gas in the combustible gas. A small number of people have studied the effects of inert gas on combustion characteristics and NO_X emission: The effects of N_2 dilution on laminar burning speed, adiabatic flame temperature, intermediate radicals and NO_X emissions of methane were investigated under different equivalence ratio conditions. Results showed that under the same equivalence ratio, the mole fraction of NO_X decreased as the N_2 doping ratio increased (Resende et al., 2019; Chu et al., 2021). The effect of CO_2 and N_2 dilution on CH_4/air flame by flame temperature distribution measurement and flame calculation was analyzed. At ambient temperature (298K), the specific heat capacity of N_2 is 1042 J/ kg·K, H_2 is 14,300 J/ kg K and that of CO_2 is 839 J/ kg K. Results indicated that CO_2 addition shows more significant effects on the thermal properties of flame, except for flame thickness (Zhang et al., 2016). Numerical simulation was used to study the combustion characteristics of laminar premixed biological combustion flames with different equivalence ratios (0.6–1.6) under standard conditions by using ANSYS CHEMKIN-PRO and GRI-MECH 3.0 mechanism. The results showed that the laminar flame speed has a good positive correlation with the adiabatic temperature under different equivalent ratios. The inhibition effect of CO_2 is stronger than N_2, which may be related to the chemical effect and heat capacity of CO_2 (Sun et al., 2021). It can be seen from the above that the change of CO_2 concentration has a stronger influence on combustion characteristics than N_2, and most scholars have studied the influence of inert gas on combustion characteristics of simple components (methane, methane-air mixture, etc.). The research on combustion characteristics and nitrogen oxide emission of complex biogas which close to actual biogas components by using simulation software is relatively rare, and further research is necessary.

Because CHEMKIN software has relatively perfect mechanism files for solving complex chemical reaction problems, it is often used to study flame combustion characteristics and NO_X conversion mechanism. It was proved to be very effective to study the combustion characteristics and NO_X emissions by using the PREMIX code of CHEMKIN(Chen et al., 2011; Gong et al., 2018). CHEMKIN and GRI-MECH 3.0 chemical kinetic models were used to simulate combustion process, which has outstanding advantages in predicting combustion chamber pressure, temperature, distribution of main combustion species and formation of nitrogen oxides (Mansha et al., 2010). Chemical dynamics simulation with hedged flame model in CHEMKIN software was carried out, and the influence mechanism of water vapor on combustion flame characteristics was analyzed, the simulated data were in good agreement with the experimental results. (Cui et al., 2020). Wilk, M. studied the combustion characteristics of carbonized biomass by TGA (Thermo Gravimetric Analyzer) analysis and CHEMKIN calculation, and the results showed that the change of gas products calculated by CHEMKIN during combustion was consistent with the results of TGA analysis (Wilk et al., 2017). The above research shows that the simulation of combustion characteristics and nitrogen oxides emission by CHEMKIN software is in good agreement with the actual situation, so it can be used to study the combustion mechanism.

The influence of one or more combustible gases on flame combustion characteristics and NO_X formation mechanism has been extensively studied, and the effects of nitrogen and carbon dioxide dilution on combustion characteristics are well explored, studies on combustion characteristics and NO_X formation mechanism of syngas flame with complex components under different inert gas concentrations are relatively scarce seldom. Therefore, in this study, typical low calorific value biogas (Li, 2019) which is closer to the real biogas composition (the composition of typical low calorific value biogas is shown in **Table 2** of **Section 2.1**) was selected to study one-dimensional laminar premixed flame, and CHENKIN software was used to simulate the influence of CO_2 concentration change on flame combustion characteristics and the formation of nitrogen oxides, aiming at providing help for the design of biomass gas burner and theoretical support for emission reduction of biomass gas boiler.

METHODS

Mechanism

In this study, based on CHEMKIN software, the premixed laminar flame characteristics and NO_X conversion mechanism of material gas were studied. All calculations in CHEMKIN software are based on mechanism files. This simulation uses two mechanism files. One is the Flame -speed-parameter-study case in CHEMKIN software, which contains C, H, O and N elements. The components include all the components in the biogas, which can well simulate the combustion process of the selected biogas. Since the generation of NO_X is not involved in this mechanism document, GRI-MECH3.0 methane mechanism document is selected for the concentration variation law of NO and NO_2. The above two mechanism documents can be used to complete the combustion characteristics of the flame and NO_X conversion mechanism.

Assuming that the gas inlet is a one-dimensional steady flow, the governing equation can be simplified as follows (Kuo, 2005; Sun et al., 2021):

Continuity

$$\dot{M} = \rho u A \tag{1}$$

In the above formula, $\dot{M}$ represents the total mass flow rate of gas components, ρmeans the density of the mixture, u represents

TABLE 1 | The main gas components of different types of biomass.

CO	H_2	CH_4	N_2	CO_2
%	%	%	%	%
6.56	14.18	15.7	50.99	3.96
8.29	13.16	14.76	46.16	7.24
20.62	5.37	5.79	47.33	15.52
16.67	8.96	9.77	42.92	21.68
25	27	3.6	0.27	23
28	30	10	5	27

the speed of the mixed fluid in the x direction, and A indicates the cross-sectional area of the flow tube.

Energy

$$\dot{M}\frac{dT}{dx}-\frac{1}{C_p}\frac{d}{dx}\left(\lambda A\frac{dT}{dx}\right)+\frac{A}{C_p}\sum_{k=1}^{k}\rho Y_k V_k C_{pk}\frac{dT}{dx}+\frac{A}{C_p}\sum_{k=1}^{k}\dot{\omega}_k h_k W_k+\frac{A}{C_p}\dot{Q}_{rad}=0 \quad (2)$$

where x represents the one-dimensional coordinates, T represents the temperature, λ is the heat conductivity of the gas, C_p represents the heat capacity of the mixed gas at constant pressure, C_{pk} expresses the kth species heat capacity at constant pressure, Y_k denotes the kth species mass fraction, V_k indicates the diffusion rate of the kth gas, h_k indicates the specific enthalpy of the kth gas, W_k represents molecular weight of the kth species, $\dot{\omega}_k$ is the production molar rate through the chemical reaction of the kth species every unit volume, and $\dot{Q}_{rad}$ expresses the radiation heat loss of mixture.

Species

$$\dot{M}\frac{dY_k}{dx}+\frac{d}{dx}\left(\rho A Y_k V_k\right)-A\dot{\omega}_k W_k=0,\left(k=1,\cdots,K_g\right) \quad (3)$$

Equation of state

$$\rho=\frac{P\bar{W}}{RT} \quad (4)$$

In **Eq. 4**, P represents the pressure in the x direction, R is the general constant of gas, and $\bar{W}$ represents the average molecular weight of the mixture.

Material

Table 1 shows the main gas components of different types of biomass gasification processes in the references (Wu et al., 2009; Kim et al., 2011; Miao et al., 2013; Wang et al., 2015). It can be noticed that the variation range of CO_2 concentration in biogas is 0–30%. In addition, related reference also showed that when the proportion of CO_2 is about 40% (Li, 2019) the fuel has better combustion characteristics and low NO_X generation. It can be seen that the influence range of CO_2 concentration on combustion characteristics and NO_X is about 0–40%, so we choose the biogas synthesized in the laboratory, that is, the typical low calorific value biogas (the composition of typical low calorific value biogas is shown in **Table 2**) as the control group for numerical simulation, and study it in the range of CO_2 concentration of 0–41.6%.

TABLE 2 | Composition of typical low calorific value biogas.

CO	H_2	CH_4	N_2	CO_2	Calorific value of gas
%	%	%	%	%	kcal/Nm^3
9.35	13.05	7.5	36.5	33.6	1260

TABLE 3 | The biogas composition under different CO_2 volume ratios.

CO	H_2	CH4	N_2	CO_2
%	%	%	%	%
14.08	19.65	11.30	54.97	0
12.73	17.76	10.22	49.69	9.60
11.60	16.19	9.31	45.30	17.60
10.48	14.62	8.41	40.89	25.60
9.35	13.05	7.50	36.50	33.60
8.22	11.48	6.60	32.10	41.60

Calculation Conditions

To ensure that the flame is close to the real burning condition, the flame speed calculator simulates a freely propagating flame at 298 K and 1 atm. At the equivalent ratio of 1.0, the laminar flame is the fastest and the flame is stable (Liu et al., 2020), so the simulation is performed at the equivalent ratio of 1.0. The initial flow rate parameter of the biogas was set to 50 cm/ s. In the simulation process, the proportion of other gases remains unchanged, only the concentration of CO_2 to be studied will be changed. **Table 3** shows the biogas composition under different CO_2 volume ratios.

RESULTS AND DISCUSSION

Effect of CO_2 Concentration on Flame Combustion Temperature

CO_2 has a high specific heat capacity and a relatively high heat absorption capacity, which has a certain influence on the flame combustion temperature, and the flame combustion temperature will have an influence on the formation of some temperature-sensitive intermediate products (Zhang, 2020). Reasonable control of the combustion temperature ensures that the fuel is fully burned, and the generation of intermediate products can be controlled to reduce the formation of pollutants while ensuring combustion efficiency.

Figure 1 illustrates the variation curve of flame temperature with distance for different CO_2 compositions simulated by CHEMKIN. For typical low calorific value biogas, when the volume ratio of CO_2 is 33.60%, it can be used as a control group to compare the influence of CO_2 concentration change on flame combustion temperature. It can be seen from **Figure 1** that with the increase of the distance and the continuation of the reaction, the temperature of the flame keeps rising. When the combustion reaches chemical equilibrium, the final temperature of the reaction keeps decreasing with the increase of CO_2 concentration. As the percentage of CO_2 increases from 0 to 41.6%, the flame temperature decreases by about 395 K, which can be explained by the fact that the concentration of small molecular intermediates such as H, O and OH in the flame decreases as the

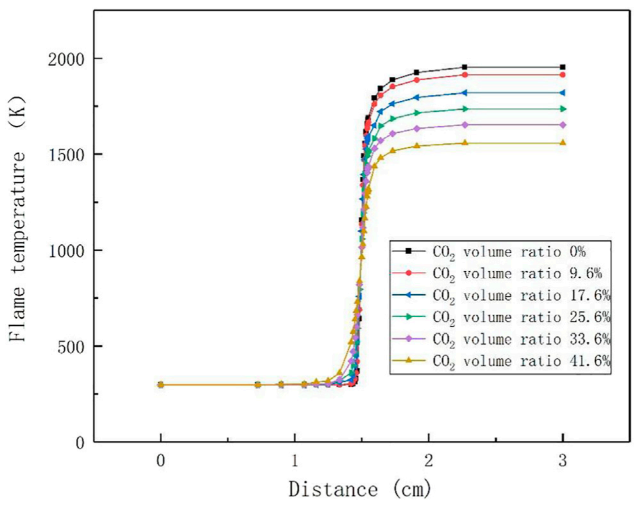

FIGURE 1 | Curve of flame temperature with distance under different CO_2 concentrations.

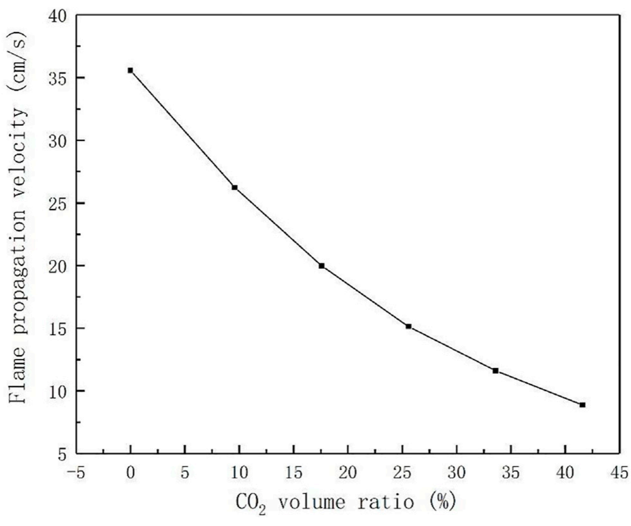

FIGURE 2 | Variation curve of flame propagation velocity with CO_2 in chemical equilibrium.

concentration of CO_2 increases, and the maximum flame temperature falls with the increase of CO_2, while the high temperature area also decreases (Cao and Zhu, 2012). The research results of Park et al. showed that with the increase of CO_2 concentration from 0 to 0.4, the flame burning temperature decreases by about 400K (Park et al., 2007), which is highly consistent with our simulation results. The effect of CO_2 concentration on combustion temperature provides a theoretical basis for adding CO_2 as a diluent to the fuel.

Effect of CO_2 Concentration on Flame Propagation Speed

The flame propagation speed of laminar flow can fully reflect the fuel activity, the basic characteristics of heat and mass transfer of combustion reaction, and the tempering characteristics of flame, etc (De Goey et al., 2011). However, the addition of CO_2 has a great influence on the flame propagation speed, so studying the influence of CO_2 concentration on the flame propagation speed can provide great help to the design of burner. Therefore, the flame propagation speed was simulated numerically using CHEMKIN software, and the variation curve of flame propagation speed with CO_2 concentration was obtained as shown in **Figure 2**. It can be seen that the flame propagation speed decreases with the increase of the volume ratio of CO_2, the flame propagation speed at 40% CO_2 concentration is 64.2% lower than that at 10%, which is consistent with the results of Hinton and Cohe (Cohe et al., 2009; Hinton and Stone, 2014). In addition, the simulation results are in good agreement with the experimental results of Maria Mitu (Mitu et al., 2017). With the increase of CO_2 concentration, the laminar flame combustion velocity decreases from the initial 35 cm/s to about 10 cm/s.

The reason why the existence of CO_2 reduces the flame propagation speed is that although CO_2 is an inert gas, it also participates in the primitive reaction $CO_2+H \rightarrow CO + OH$(Glarborg and Bentzen, 2008), which inhibits the oxidation of CO. It will also compete with the branch reaction of the main chain for H element, leading to the reduction of H group and OH group (Huang, 2011), and these two free radicals have a great promotion effect on the flame propagation speed. Consequently, the volume ratio of CO_2 in syngas should be controlled to ensure a reasonable combustion rate.

Effect of CO_2 Concentration on the Formation of NO and NO_2

The emission of nitrogen oxides is a problem to be considered in small-capacity boilers, because the nitrogen oxides produced by any fossil fuel combustion will lead to the formation of acid rain and photochemical smog, which are important causes of air pollution (Werle, 2015). Thus, studying the effect of CO_2 concentration on NO_X formation can reduce the pollutant generation at the source and can provide theoretical support for reducing NO_X emissions for biomass gas boilers.

Figure 3A shows the curve of NO mole fraction with distance under different CO_2 concentration, and **Figure 3B** presents the variation curve of NO mole fraction with CO_2 concentration in chemical equilibrium. From the above two figures, it can be observed that the molar fraction of NO keeps increasing with the distance and the reaction proceeds continuously. When the biogas is devoid of CO_2, the molar fraction of NO increases fastest with distance, and when the combustion reaches chemical equilibrium, the proportion of NO produced is the largest, and the molar fraction of NO produced at equilibrium declines as the volume proportion of CO_2 in the fuel increases. This is in agreement with the findings of Tang (Tang et al., 2009). The influence of CO_2 concentration on NO generation was studied by using a tube furnace with Li of 1373K and equivalent ratio of 1. The experimental results showed that with the increase of CO_2 concentration, NO concentration decreases from 14.8 to 5.9 ppm (Li, 2019), which is consistent with our simulation results.

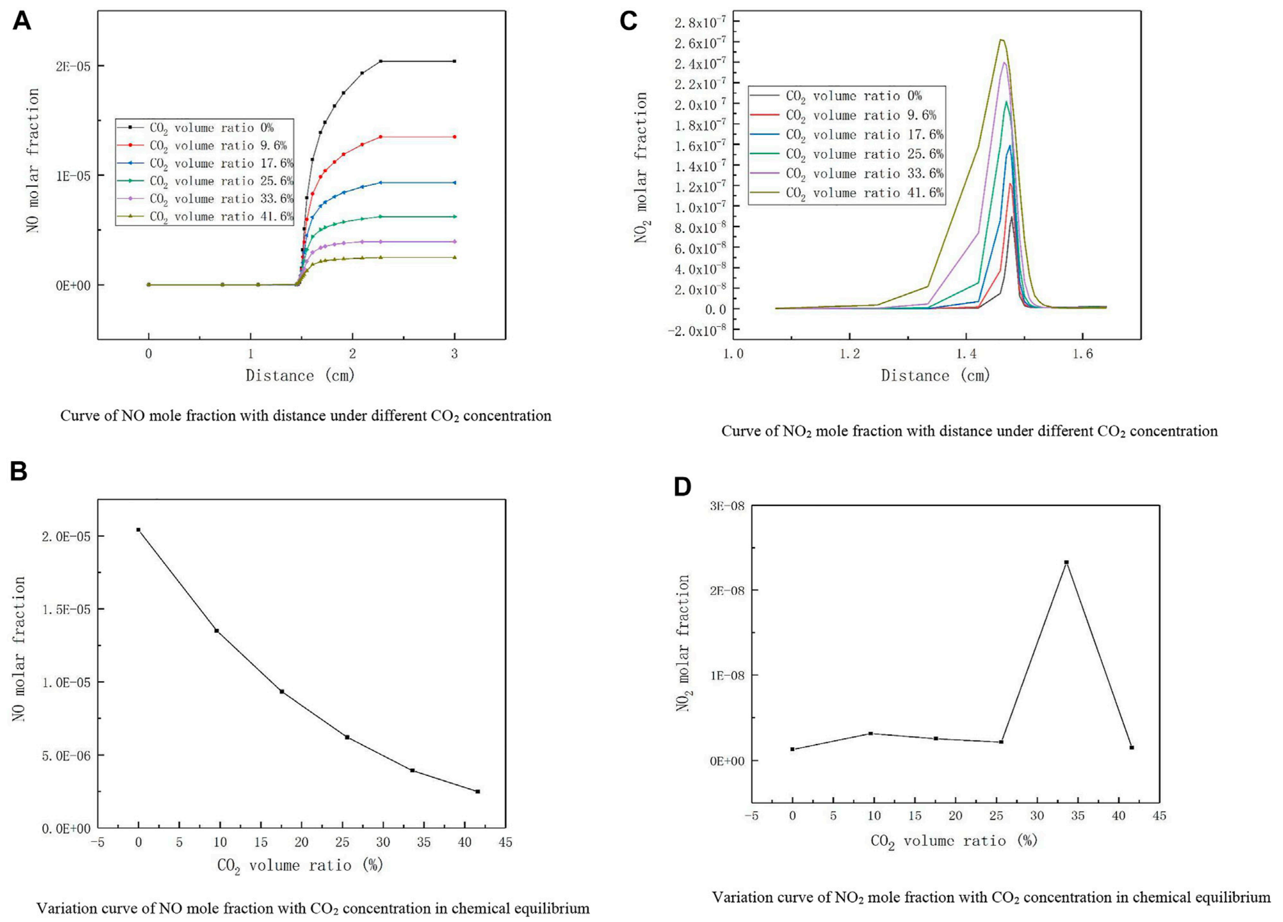

FIGURE 3 | Distribution curve of nitrogen oxides under different CO_2 concentrations. **(A)** Curve of NO mole fraction with distance under different CO2 concentration. **(B)** Variation curve of NO mole fraction with CO2 concentration in chemical equilibrium. **(C)** Curve of NO2 mole fraction with distance under different CO2 concentration. **(D)** Variation curve of NO2 mole fraction with CO2 concentration in chemical equilibrium.

Because of the different gas composition and the influence of experimental operation process, the specific values are different. Liu and Dong et al. demonstrated that CO_2+H→CO + OH and CO_2+CH→HCO + CO are the main chemical reaction in which CO_2 participates, while these two reactions indirectly affect the production of the NO precursor HCN, thus inhibiting the production of NO_X (Liu et al., 2001; Dong et al., 2014).

Figure 3C shows the curve of NO_2 mole fraction with distance under different CO_2 concentration, and **Figure 3D** presents the variation curve of NO_2 mole fraction with CO_2 concentration in chemical equilibrium. From **Figure 3C**, it can be found that the molar fraction of NO_2 at the peak is highest when the volumetric proportion of CO_2 in the biogas is 41.6%, with a value of 2.06E-07. As the volumetric proportion of CO_2 in the biogas decreases, the molar fraction of NO_2 at the peak also reduces. As can be noticed from **Figure 3D**, the molar fraction of NO_2 produced at equilibrium is maximum for the selected typical low calorific value biogas whose CO_2 concentration is around 33.6%, and the molar fraction of NO_2 produced is significantly lower in other cases. Thus, it can be seen that the effect of the variation of CO_2 concentration on NO_2 is very complex. This may be due to the fact that the concentration of H, O, and HO_2 changes with different CO_2 concentrations, leading to unstable formation of NO_2(Glarborg and Bentzen, 2008). Since the combustion mechanism is very complex the exact cause needs to be further verified.

CONCLUSIONS

In this paper, the influence of CO_2 concentration on flame combustion characteristics and NO_X formation was studied by using CHEMKIN software. The main research results are as follows.

(1) With the increase of CO_2 concentration, the flame combustion temperature and flame propagation speed gradually decrease, and the flame propagation speed decreases even more slowly. Compared with syngas without CO_2, when the CO_2 concentration in biogas is 41.6%, the flame temperature is 395 K lower and the flame propagation speed decreases by 64.2%. It can be seen that the existence of CO_2 in biogas not only reduces the combustion temperature of the flame, but also greatly reduces the propagation speed of the laminar flame.

(2) After the combustion reaches the chemical equilibrium, the molar fraction of NO in the product decreases with the increase of CO_2, while the molar fraction of NO_2 does not show regularity with the change of CO_2 concentration.When the concentration of carbon dioxide is 33.6%, that is, when the research object is typical low calorific value biogas, the mole fraction of nitrogen dioxide in chemical equilibrium is the highest.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

JM performed the data analysis and wrote the manuscript. CQ performed the simulations using software. SL provided ideas and directed the analysis of the data. ZZ provided constructive comments on revisions to the manuscript.

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