



Influence of H₂O on Oxygen Enriched Diffusion Combustion of Natural Gas

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ABSTRACT

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O₂/H₂O combustion technology, as the next generation of oxy-fuel combustion technology with great potential, can greatly increase the utilization rate of clean energy CH₄. In this paper, the natural gas combustion process under 6 operating conditions of O₂/H₂O atmosphere and O₂/FH₂O atmosphere is numerically simulated. The horizontal analysis is carried out on the characteristics of H₂O fraction, CO₂ volume fraction and the amount of pollutants (NO_x, carbon black), and in-depth exploration of the content of additive H₂O and the influence of chemical action on the above characteristics. The research results show that the chemical effects of H₂O have a negative effect on combustion temperature, and the physical effects are dominant. The chemical effects of H₂O have a great impact on CO production and little effect on the production of CO when the proportion of H₂O is 65-79%. The chemical effects of H₂O inhibit the formation of NO_x and carbon black when the proportion of H₂O is within the range of 55-70%. The chemical effect has the greatest impact on the formation of dyes (NO_x, carbon black) when the proportion of H₂O is within the range of 65-70%.

1. INTRODUCTION

As one of the most widely used technologies in modern industrial combustion, oxy-fuel combustion technology has low risk and high feasibility and high efficiency. The new O₂/H₂O oxy-fuel combustion technology can greatly increase the utilization rate of clean energy CH₄ [1-3], and its combination with the production process of natural gas hydrate can effectively promote the development of natural gas industry, and alleviate global warming and environmental pollution from the source.

As the next generation of oxy-fuel combustion technology with great potential, O₂/H₂O combustion technology does not need to recycle the flue gas, and the combustion temperature adjustment is achieved by using water vapor to participate in the combustion process. This new type of oxy-fuel combustion technology can make the entire combustion system more concise and compact, and the start-stop process is easier. It not only reduces the system cost [4-6], but also makes full use of the waste heat of the flue gas, thus reducing the loss of waste heat, and avoiding the problem of impurity gas enrichment in O₂/CO₂ flue gas cycle more effectively [7-10]. Scholars at home and abroad have conducted extensive theoretical and experimental research on oxy-fuel combustion technology, most of which are related to the combustion characteristics, pollutants (NO_x, carbon black) of oxy-fuel combustion in O₂/N₂, O₂/CO₂ atmospheres and the basic research work on the physicochemical effects of additives, but lack of application research of O₂/H₂O combustion technology. O₂/H₂O combustion technology has its unique advantages in terms of combustion characteristics and pollutant emissions. Therefore, a full understanding of the effect of additive H₂O on oxygen-enriched diffusion combustion is of great significance to the

development of O₂/H₂O oxy-fuel combustion technology [11].

Therefore, it is necessary to analyze the different working conditions under O₂/H₂O atmosphere to have a clearer understanding of O₂/H₂O combustion technology and the influence of H₂O on oxygen enriched diffusion combustion of natural gas, and to provide more sufficient basic theoretical support for the industrial application of O₂/H₂O combustion technology in the future. Thus, this paper analyzes the distribution of combustion temperature and pollutant (NO_x, carbon black) generation under the same O₂ concentration and different H₂O concentration conditions, and explores the influence characteristics of H₂O content on combustion temperature, CO volume fraction, CO₂ volume fraction and pollutant (NO_x, carbon black) concentration. By analyzing the differences of combustion temperature, CO volume fraction, CO₂ volume fraction and pollutant (NO_x, carbon black) generation under different conditions of O₂/H₂O atmosphere and O₂/FH₂O atmosphere with the same concentration proportion, the influence of chemical action of H₂O additive on oxygen enriched diffusion combustion was studied. Furthermore, the optimal concentration range of O₂ and H₂O in oxidant was determined by overall comparative analysis.

2. NUMERICAL SIMULATION METHOD AND CONTROL EQUATION

2.1 Mathematical model

In the process of numerical simulation, the standard model and the general finite rate model are selected as the turbulent flow model and the combustion model respectively; each radiation heat transfer model needs to be comprehensively

considered in the applicable occasions, calculation amount, calculation accuracy, and hardware CPU performance requirements. Thus, the p-1 model is defined as the radiation heat transfer model; the probability density function (pdf) model is used as a mathematical model of the nitrogen oxide pollutant reaction mechanism and information transmission process, and the NO concentration transport equation provided by Fluent is used to predict the emission of NOx and pollutants.

$$\frac{\partial}{\partial t}(\rho Y_{NO}) + \nabla \cdot (\rho \bar{v} Y_{NO}) = \nabla \cdot (\rho D \nabla Y_{NO}) + S_{NO} Y_{NO} \quad (1)$$

In the above formula, ρ is the density, Y_{NO} is the volume fraction of NO, D is the diffusion coefficient, S_{NO} is the generation rate of NO, t is the time, \bar{v} is the average rate;

The single-step Khan and Greeves model is selected for the carbon black generation model. In terms of the gas phase chemical reaction mechanism, it adopts the same four-step simplified reaction mechanism as the combustion form of CH₄. Separation solver, implicit format, finite volume method, and SIMPLE algorithm are used to solve the calculation.

2.2 Geometric model

2.2.1 Model simplification

In this paper, the cylindrical burner is used in the simulation, and the geometric model is established by using fluent preprocessing software gambit. The simplified geometric shape and size are shown in Figure 1. It can be clearly seen from the figure that the whole length of the burner is 3 m, the radius is 0.3m, and there is a nozzle with a radius of 0.006m in the middle. The fuel methane enters the burner along the nozzle, and the oxidant enters the annular area of the combustion chamber inlet (excluding the nozzle), mixes with methane and burns, diffuses into the combustion chamber at the same time, and finally produces turbulent diffusion flame. In order to obtain a clearer diffusion flame, an outer wall is installed to separate the flammable methane and oxidizer, and the length of the outer wall is specified as 0.05m.

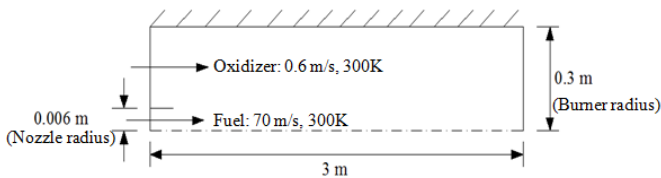


Figure 1. Schematic diagram of burner structure

2.2.2 Grid division

After the model is built, perform numerical calculation and mesh division. The detailed steps are to divide the spatially continuous solution area into multiple small sub-areas, and confirm each node separately. There are many factors that need to be considered in the division of the grid, so the workload will be relatively large. The grid format (such as structured and unstructured grid) of the division can directly affect the accuracy and complexity of the calculation results. Generally, the accuracy of the calculation result can be judged by the number of grids: the more grids, the higher the accuracy of calculation, but the calculation efficiency is low; the number of grids is small, the calculation accuracy is low and even the convergence cannot be achieved. Thus, the correct

and reasonable grid division is the prerequisite for ensuring high-quality grids.

Gambit software is used to divide the overall grid of the solution area, and the grid division diagram is shown in Figure 2. Considering that solving the regional data structure is relatively simple, a quadrilateral structured grid is adopted to ensure the formation of high-quality grids in a short time. The overall presentation is graded grid, that is, along the axial gas flow, the grid is increasingly sparse, while considering a number of important areas have great impacts on the calculation results, such as gas inlet, exterior in the vicinity and inside the burner, the divided grids need to be encrypted. A total of 20300 nodes and 20075 grid cells are divided.

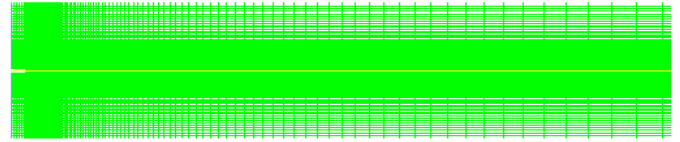
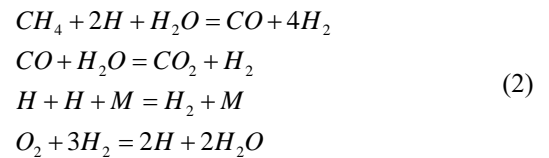


Figure 2. Grid division diagram

2.3 The mechanism of gas phase chemical reaction and solving method

(1) To simplify the calculation complexity and improve the efficiency, the four-step simplified chemical reaction mechanism of methane combustion is adopted:



(2) The solver used in the simulation is a separate pressure-based separation solver, which solves the control equations in sequence. In the process of discretizing nonlinear partial differential equations, to directly transform the nonlinear partial differential equations into linear algebraic equations on a grid element, the finite volume method can be considered. We can choose the standard format as the pressure interpolation format. The momentum equation, the turbulence kinetic energy equation, the turbulence dissipation rate equation, the component mass conservation equation and the energy equation can all be selected as the first-order upwind scheme, because it has the remarkable characteristics and advantages of the second-order upwind scheme such as high stability and fast calculation speed. SIMPLE algorithm is widely used in the actual research and calculation of fluid mechanics and computational heat transfer, and its sub-relaxation factor is a numerical calculation method that can be widely used to calculate the flow of objects at any velocity, and the range of sub-relaxation factor can be expanded in an appropriate amount. Accordingly, this paper will use the SIMPLE algorithm to solve the pressure-velocity coupling scheme. In the setting of the residual value, the residual value of the energy equation and the P-1 radiation heat transfer equation is set to 10^{-6} , and the other governing equations are all set to 10^{-4} . In setting the sub-relaxation factor, the sub-relaxation factors of pressure, momentum, k and ϵ are set to 0.3, 0.7, 0.8 and 0.8 respectively.

2.4 Governing equation

Establishing the governing equation is the first step of numerical simulation. A general form of the governing equation is established to facilitate the analysis of each governing equation [12-16]:

$$\frac{\partial}{\partial x}(\rho u \varphi) + \frac{\partial}{r \partial r}(r \rho v \varphi) + \frac{\partial}{r \partial \theta}(\rho w \theta) = \frac{\partial}{\partial x} \left[\Gamma_{\varphi} \frac{\partial \varphi}{\partial x} \right] + \frac{\partial}{r \partial r} \left[r \Gamma_{\varphi} \frac{\partial \varphi}{\partial r} \right] + \frac{\partial}{r^2 \partial \theta} \left[\Gamma_{\varphi} \frac{\partial \varphi}{\partial \theta} \right] + s_{\varphi} \quad (3)$$

In the above equation, φ is the universal dependent variable, Γ_{φ} is the transport coefficient, S_{φ} is the source term, ρ is the density, x, r, θ respectively represent the axial coordinates, radial coordinates, circumferential coordinates, u, v, w respectively represent the component of the velocity vector in the x, r, θ direction. (1) When $\varphi=1, \Gamma_{\varphi}=0$, it represents the continuity equation; (2) When $\varphi=u, v, w$, it represents the momentum equation corresponding to the direction; (3) When $\varphi=k$, it represents the turbulent energy equation; (4) When $\varphi=\varepsilon$, it represents the turbulent energy dissipation rate equation; (5) When $\varphi=f$, it represents the equation of conservation of component mass; (6) When $\varphi=h$, it represents the energy equation. Where k is the turbulent kinetic energy, ε is the turbulent energy dissipation rate, f is the mass fraction, and h is the enthalpy.

3. BOUNDARY CONDITIONS AND CALCULATION CONDITIONS

3.1 Boundary conditions

The fuel methane nozzle was set as the velocity inlet

boundary, and the velocity was 70m/s. In this case, the turbulence intensity and hydraulic diameter were set at 10% and 0.006m respectively, and the temperature was set at 300K. The velocity inlet boundary was set at the oxidant inlet. When H₂O was added to the oxidant, it was necessary to ensure that the content of O₂ added each time remained unchanged, while the content of the additive H₂O gradually increased (or decreased). Taking the working condition CH₄/21% O₂/79% H₂O and the flow rate of the oxidant 0.6m/s as the benchmark. The flow rates of the oxidant in the set conditions were successively calculated as 0.42m/s, 0.36m/s, 0.315m/s, 0.28m/s and 0.252m/s. The turbulence intensity and hydraulic diameter were set at 10% and 0.294m, respectively, and the temperature was set at 300K. The outlet of the burner was set as the pressure outlet boundary, the gauge pressure was set as 0, and the turbulence intensity and hydraulic diameter were set as 10% and 0.3m respectively. At the same time, the backflow at the pressure outlet should also be considered. The other boundaries were set as wall boundaries, and since burner walls usually have cooling devices, the wall temperature was specified as 300K.

3.2 Calculation conditions

In view of the particularity of the chemically active substance H₂O, it is necessary to consider its chemical action as the key point. Thus, a fictional substance FH₂O is set up, which has the same thermal properties, transport properties and radiation properties as H₂O, but does not participate in chemical reactions.

Six working conditions (21%O₂, 30%O₂, 35%O₂, 40%O₂, 45%O₂, 50%O₂) were selected for the numerical simulation of natural gas oxygen-enriched diffusion combustion in the O₂/H₂O atmosphere. FH₂O was added to the oxidant instead of H₂O, and the same boundary conditions and working conditions were set. The specific calculation working conditions are shown in Table 1.

Table 1. Calculation conditions

Calculation conditions		Gas species	Velocity, m/s	
fuel		CH ₄	70	
	case1	21% O ₂ /79% H ₂ O	0.6	
O ₂ /H ₂ O atmosphere	case2	30% O ₂ /70% H ₂ O	0.42	
	case3	35% O ₂ /65% H ₂ O	0.36	
	case4	40% O ₂ /60% H ₂ O	0.315	
	case5	45% O ₂ /55% H ₂ O	0.28	
	case6	50% O ₂ /50% H ₂ O	0.252	
	oxidizer	case7	21% O ₂ /79% FH ₂ O	0.6
case8		30% O ₂ /70% FH ₂ O	0.42	
case9		35% O ₂ /65% FH ₂ O	0.36	
O ₂ /FH ₂ Oatmosphere		case10	40% O ₂ /60% FH ₂ O	0.315
case11		45% O ₂ /55% FH ₂ O	0.28	
case12		50% O ₂ /50% FH ₂ O	0.252	

4. RESULTS AND DISCUSSION

4.1 Effect of additive H₂O content on oxy-fuel diffusion combustion

Figure 3 shows the distribution curve of combustion temperature on the central axis under 6 working conditions.

As shown in the figure, the combustion temperature gradually decreases with the increase of the content of additive

H₂O. In the axial direction, the effect of H₂O addition on combustion temperature increases first and then decreases. According to calculation, the maximum combustion temperature under six working conditions is 1517.4K, 1838.7K, 2261.8K, 2409.3K, 2622.8K and 2752.9K respectively. As shown in Figure 4, when the proportion of additive H₂O is in the range of 65%-70%, the decrease rate of the maximum combustion temperature is the fastest, which indicates that the addition of H₂O in this concentration range

has the greatest impact on the combustion temperature in the whole burner.

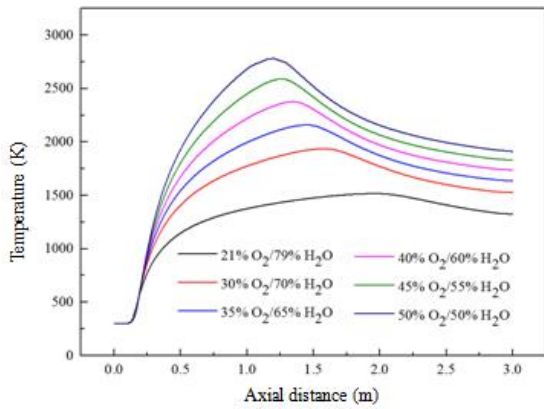


Figure 3. Distribution of combustion temperature on the central axis

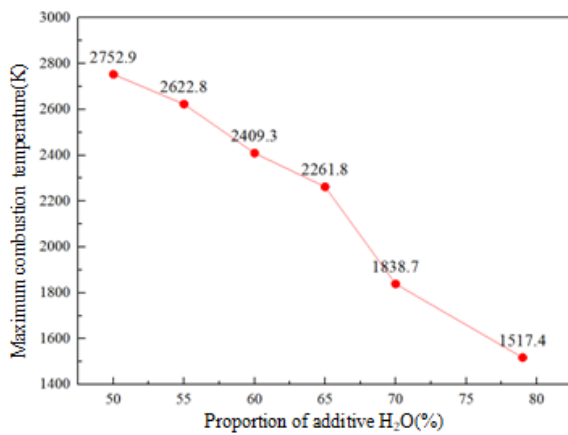


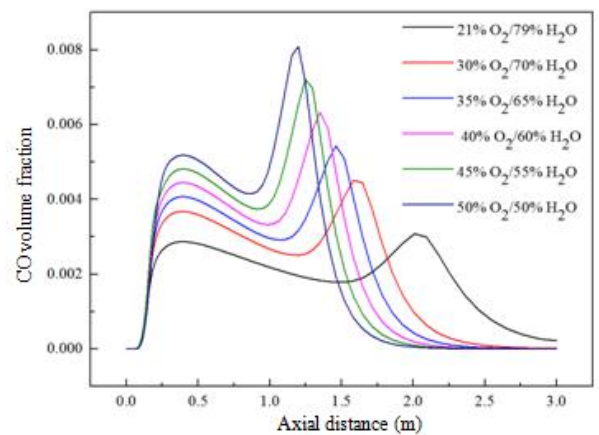
Figure 4. Maximum combustion temperature in six operating conditions

As products in the combustion process, the production of CO and CO₂ will inevitably be affected by the concentration of reactants H₂O. Figure 5 shows the distribution curves of CO volume fraction and CO₂ volume fraction on the central axis under 6 working conditions. As shown in Figure 5(a), within the zones Z=0-0.25m and Z=2.5-3.0m, CO production remained almost unchanged with the increase of additive H₂O content; within the zones Z=0.25-1.5m, CO production decreased with the increase of additive H₂O content; within the zones Z=1.5-2.5m, with the increase of the content of additive H₂O, the amount of CO production also increases. As shown in Figure 5(b), CO₂ production decreases with the increase of the content of additive H₂O, that is, the addition of H₂O inhibits CO₂ generation. In the axial direction, the influence degree of H₂O addition on CO₂ production shows a trend of first increasing, then decreasing and then increasing.

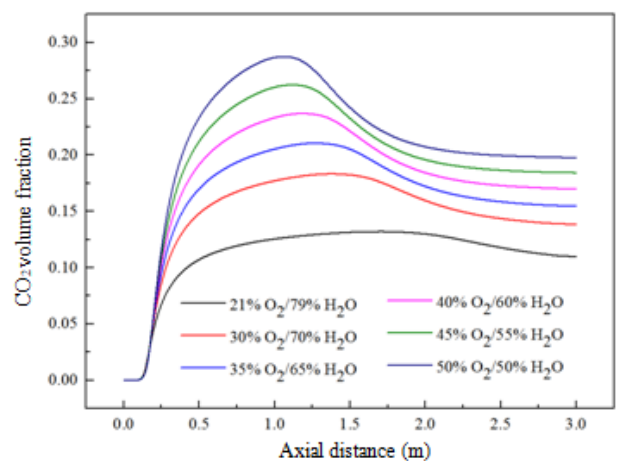
Figure 6 shows the distribution curve of NO_x volume fraction on the central axis under 6 working conditions. As shown in the figure, the volume fraction of NO_x in the condition of 21%O₂/79%H₂O spikes in the axial direction, which may be due to the combustion temperature below 1600K in the condition. The factor affecting the generation of NO_x is the length of time that the reactants stay in the high temperature zone, while the high concentration of H₂O in the oxidizer does not participate in the reaction at the beginning.

As a result, the production of NO_x will increase rapidly with the extension of retention time before the combustion reaction reaches chemical equilibrium [17-20]. The overall analysis of the other 5 working conditions shows that the NO_x production decreases with the increase of the content of H₂O additive. And when the proportion of additive H₂O is in the range of 55%-60%, the addition of H₂O has the greatest influence on NO_x generation. In the axial direction, the effect of H₂O addition on NO_x generation increases at first and then decreases [21].

Figure 7 shows the distribution curve of carbon black volume fraction on the central axis under 6 working conditions. According to the figure, within the zone Z=0-0.25m, the amount of carbon black production almost remained unchanged with the increase of the additive H₂O content; within the zone Z=0.25-1.5m, the amount of carbon black production decreased with the increase of the additive H₂O content; within the zone Z=1.5-3.0m, the amount of carbon black production decreased with the increase of the additive H₂O content. In the axial direction, the effect of the addition of H₂O on the carbon black production increased at first and then decreased until it was almost unaffected. As shown in Figure 8, the change curve of the maximum volume fraction of pollutant NO_x and carbon black with the increase of additive H₂O content shows that, when the proportion of additive H₂O is within the range of 55%-60%, the addition of H₂O inhibits the generation of pollutants in the whole burner, and the inhibition effect is the largest.



(a) CO Volume fraction



(b) CO₂ Volume fraction

Figure 5. Volume fraction distribution on the central axis

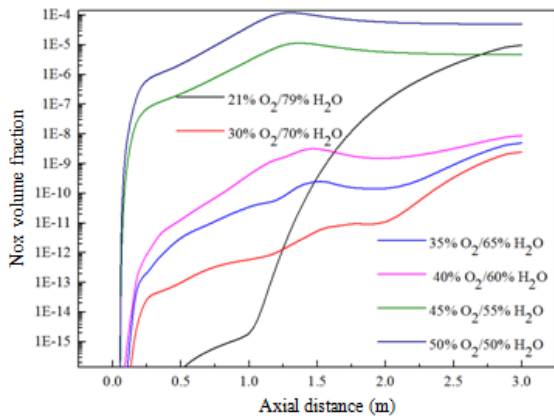


Figure 6. Distribution of NO_x volume fraction on the central axis

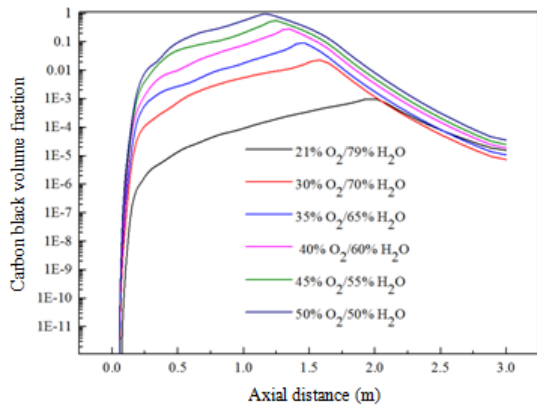


Figure 7. Distribution of soot volume fraction on the central axis

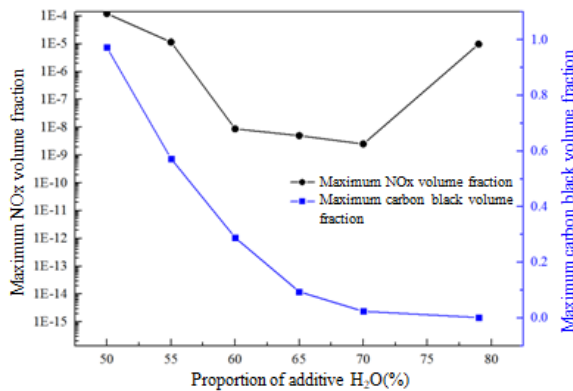


Figure 8. Volume fraction of maximum pollutants (NO_x, soot) in six operating conditions

4.2 Effect of chemical action of additive H₂O on oxy-fuel diffusion combustion

In order to distinguish the influence of chemical action on oxygen-rich diffusion combustion, the simulation results of six working conditions in O₂/H₂O atmosphere were compared with those in O₂/FH₂O atmosphere, and the difference between the two was caused by chemical action. Figures 9 and 10 are the comparison of the maximum combustion temperature under two atmospheres and the fitting curve of temperature difference respectively. The temperature differences under conditions of 50%H₂O, 55%H₂O, 60%H₂O, 65%H₂O,

70%H₂O and 79%H₂O were calculated as 193.4K, 259.6K, 176.7K, 121.5K, 61.3K and 110.8K, indicating that when high concentration of H₂O is added to the oxidizer, in the process of CH₄ oxy-fuel diffusion combustion, there will be chemical action, which has a negative impact on combustion temperature. With the increase of additive H₂O content, the temperature difference decreases first, then gradually increases, and then decreases. The chemical action has the greatest impact on combustion temperature when the proportion of additive H₂O is in the range of 65%-79%.

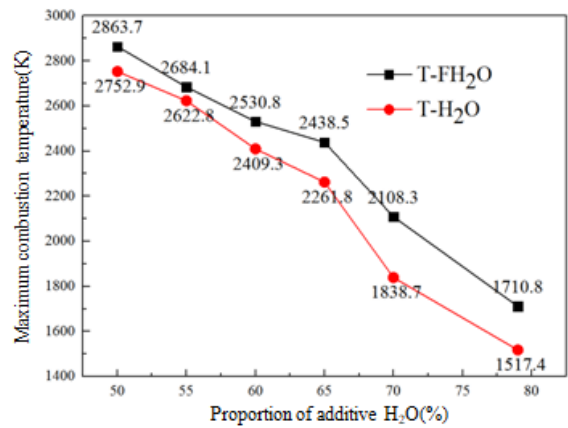


Figure 9. Comparison of the highest combustion temperatures

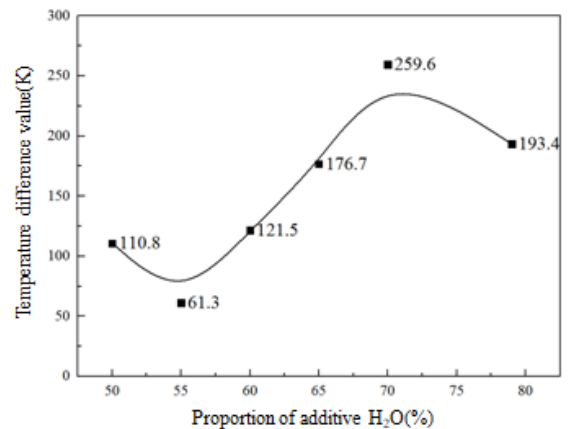
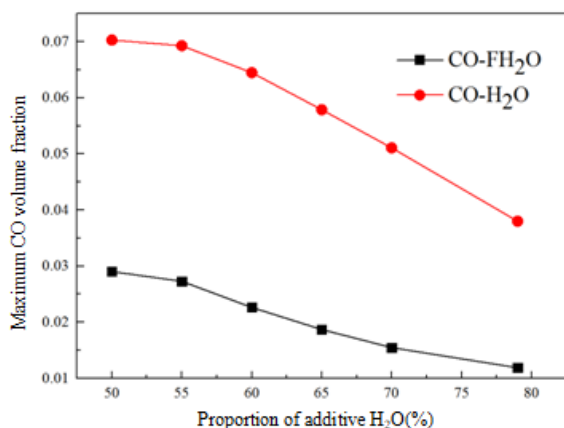


Figure 10. Fitting the temperature difference

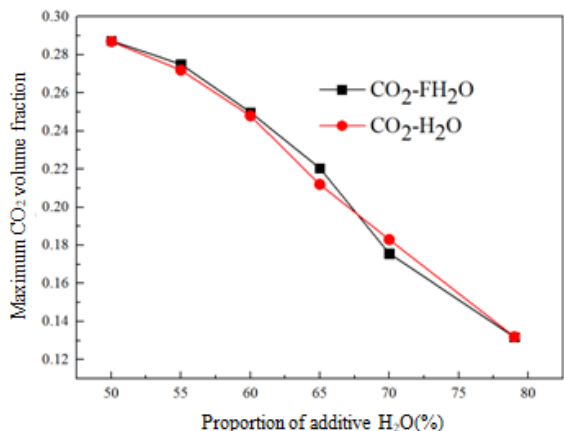
Figure 11 shows the contrast curves of the maximum CO volume fraction and the maximum CO₂ volume fraction under the O₂/H₂O atmosphere and the O₂/FH₂O atmosphere respectively. The comparison shows that the chemical effect of H₂O has a great influence on the generation of CO, and can promote the generation of CO to a large extent in the reaction process. On the whole, the chemical effect of H₂O has little effect on the generation of CO₂, and it can be considered that it is completely affected by physical effects, and the chemical effect can be ignored. More specifically, it has a slight inhibition effect on CO₂ generation when the additive H₂O proportion is in the range of 50%-65%, and can slightly promote CO₂ generation when the additive H₂O ratio is in the range of 65%-79%.

Figure 12 shows the contrast curves of the maximum NO_x volume fraction and the maximum carbon black volume fraction at the outlet under six working conditions under two atmospheres. As shown in Figure 12(a), the chemical effect of

H₂O promotes the generation of NO_x when the content of H₂O is in the range of 50%-55% and 70%-79%; the chemical effect of H₂O has an inhibitory effect on the generation of NO_x, and the inhibitory effect is the most obvious when the content of H₂O is in the range of 55%-70%. As shown in Figure 12(b), the chemical action of H₂O also inhibits the generation of carbon black, and the influence is greatest when the content of H₂O is in the range of 65%-70%. In general, the addition of H₂O can inhibit the generation of pollutants (NO_x, carbon black) in the combustion process when the content of H₂O is in the range of 55%-70%, and the inhibition effect on pollutants is the best when the content of H₂O is in the range of 65%-70%.



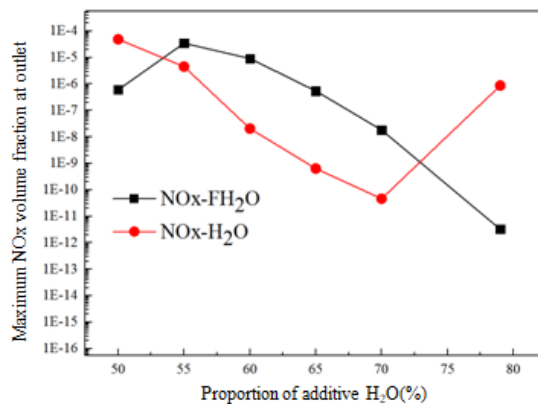
(a) CO volume fraction



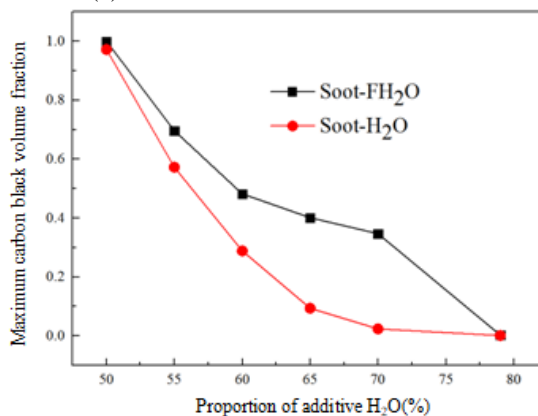
(b) CO₂ volume fraction

Figure 11. Highest volume fraction comparison

In order to more intuitively observe the effect of chemical action on methane oxy-fuel diffusion combustion degree, and compared with the physical action, determine its primary and secondary position in the reaction process. Table 2 shows the proportion of chemical inhibition on combustion temperature and pollutants (NO_x, carbon black) when the proportion of additive H₂O is 65% and 70%. In the table, φ_T , φ_{NO_x} and φ_{Soot} represent the corresponding proportions respectively. As can be seen from the table, in the inhibition effect of H₂O on combustion temperature, physical effect plays a dominant role (accounting for more than 85%), while chemical effect has a small effect (accounting for less than 10%). In the inhibition effect of H₂O on the production of pollutants (NO_x, carbon black), chemical action and physical action play an equal role (both are about 50%). In the process of oxy-fuel diffusion combustion, the generation of pollutants is inhibited at the same time.



(a) Maximum NO_x volume fraction



(b) Maximum soot volume fraction

Figure 12. Comparison of maximum volume fraction at the outlet

Table 2. Proportion of chemical inhibition

	65%H ₂ O	70%H ₂ O
φ_T	6.76%	11.34%
φ_{NO_x}	49.97%	49.73%
φ_{Soot}	43.37%	48.24%

5. CONCLUSIONS

In this paper, based on the principle of constant O₂ concentration and gradient change of H₂O concentration, the boundary conditions and calculation conditions required in the simulation were determined. The combustion process under six different conditions of O₂/H₂O atmosphere and O₂/FH₂O atmosphere were numerically simulated. From the characteristics of combustion temperature, CO volume fraction, CO₂ volume fraction and pollutant (NO_x, carbon black) production, the horizontal analysis was carried out, and the influence rules of additive H₂O content and chemical action on the above characteristics were deeply explored. The conclusions were as follows:

(1) The chemical effect of H₂O has a negative influence on the combustion temperature, and the physical action plays a dominant role. The chemical action has the greatest influence on the combustion temperature when the proportion of H₂O is in the range of 65-79%.

(2) The chemical effect of H₂O has a great influence on the generation of CO, and has a significant promotion effect on the production of CO; the chemical effect of H₂O has little effect on the generation of CO₂, which is almost negligible,

and it is completely controlled by the physical effect.

(3) The chemical effect of H₂O has a significant inhibitory effect on the formation of NO_x when the proportion of H₂O is in the range of 55%-70%; the chemical effect of H₂O inhibits the formation of carbon black; the chemical effect has the greatest influence on the formation of dyes (NO_x, carbon black) when the proportion of H₂O is in the range of 65-70%.

Based on the above research and analysis, the optimal concentration range of H₂O can be determined to be 65-79% based on the influence of H₂O content and chemical effect on combustion temperature and pollutant generation.

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NOMENCLATURE

Y	volume fraction
D	diffusion coefficient
S	generation rate

t time, s
 \bar{v} average rate, m. s⁻¹

Greek symbols

ρ density, kg. m⁻³

φ universal dependent variable
 Γ_φ transport coefficient
 x, r, θ axial coordinates, radial coordinates, circumferential coordinates
 u, v, w the component of the velocity vector in the x, r, θ direction, m. s⁻¹