Balance Calculation and Optimization of Denitrification Process Simulation of Oxy-fuel Combustion Flue Gas

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Abstract. Oxy-fuel combustion is an important development direction of combustion technology, and the key technology to reduce the cost of flue gas purification is the reduction of denitration cost. Based on the experiments and simulation process with Aspen Plus of pressure denitrification process, the reaction time and equilibrium concentration were analyzed when the denitrification reaction was balanced. Finally, the denitrification process was optimized according to the simulation results. The results showed that the equilibrium concentration and time of the denitrification process decreased with the increase of pressure, initial oxygen content and the decrease of temperature. The content of NO in flue gas had little influence on equilibrium concentration and time. After analysis and calculation, it is considered that 30°C and 26bar is more suitable for denitrification process of the pressurized flue gas.

Introduction

Oxy-fuel combustion technology is also called CO_2/O_2 cycle combustion technology. It has good continuity with traditional coal-fired power generation technology in technology and workmanship. Economic evaluation also shows that oxy-fuel combustion technology is most competitive on equipment investment cost and generation cost compared with the same kind of technology now. This combustion mode has become one of the most important development direction of coal combustion technology in this century[1,2].

In oxy-fuel combustion process, oxygen separation from air and CO_2 compression will cost greater energy consumption which will increase the cost of electricity generation greatly. Therefore, reducing the cost of power generation by various means faces a significant challenge. And reducing the cost of flue gas purification is one of the most important ways. The desulfurization cost of coal-fired power plants is low, and the cost of denitrification is very high. So reducing the denitration cost of oxy-fuel combustion is very importance to reduce the cost of flue gas purification[3,4].

For this purpose, the process of single denitrification in compression process is studied. Based on the experimental results of the pressure denitrification process, simulation process is built by using Aspen Plus. Effects of pressure, temperature, initial oxygen concentration, initial NO concentration and the presence of N_2 in flue gas on the pressure denitrification process is systematically analyzed. In this paper, the reaction time and equilibrium concentration are analyzed when the system reaction reached equilibrium. Finally, the pressure denitrification process is optimized according to the simulation results. The rationality and feasibility of this mechanism are further proved.

Model of Pressure Denitrification and Result Analysis

Firstly, the pressure denitrification experiments are carried out under some conditions on the experiment bench. The experiment advent gas is a mixture of NO, SO_2 and CO_2. The experiment conditions and results are shown in Table 1. Then according to the experiment process and conditions, the Aspen Plus process simulation is performed. The established Aspen Plus process is shown in Fig.1.

According to the above experiment equipment and conditions, a series of Rbatch modules are simulated based on the dynamics principle. Rbatch1 simulates CO_2 cleaning process. Rbatch2 simulates flue gas pressure boosting. Rbatch3 simulates constant pressure reaction process. The

export mixture carries out flash separation and dehydration at the end of the reaction[6,7]. Then the gas composition is analyzed to study the denitration rate of the denitration process.
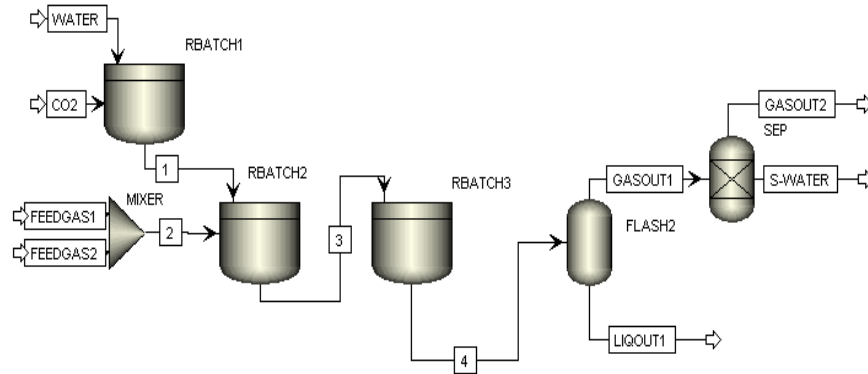

Fig.1 Flow chart of pressurized denitrification of oxy-fuel combustion flue gas

Ensured that the simulation process is the same as the flue gas and operating conditions of the experiment process, the experiment research and Aspen process simulation of pressure denitrification under different conditions are carried out in the reactor. The conditions of denitrification, the conditions and results of experiment and simulation are shown in Table 1.

Table 1. Comparison of simulation and experimental results

Initial concentration		Temperature /° C	Pressure /bar	Residence time /min	Simulated NO removal rate /%	Experimental NO removal rate /%	relative error/%
NO/ppm	O_2/%						
279	5	25	10	5	76.0	76.0	0
836	5	25	6	5	83.5	82.7	1.0
836	5	25	10	5	92.5	92.2	0.3
836	5	25	10	10	95.9	95.2	0.7
836	5	30	10	5	91.1	91.0	0.1
836	10	25	10	5	94.4	93.5	1.0
836	5	25	14	5	94.9	95.3	-0.4

As shown in Table 1, by comparing the experiment and simulation results, the NO removal rate is in good agreement with the data. The maximum relative error is only 1%. It shows that the model and mechanism used in simulation are reliable and feasible.

Balance Simulation of Pressure Denitrification Process
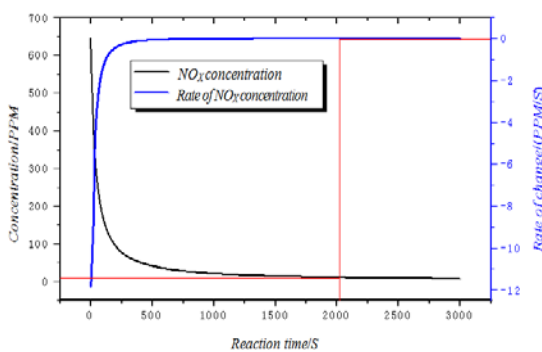

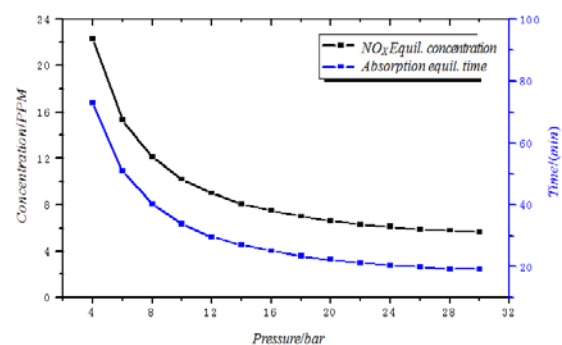Fig.2 Relationship between NOx and change rate with reaction time under 10bar

Fig.3 Effect of pressure on NO absorption equilibrium time and concentration

In order to consider the economical efficiency, when the reaction reaches equilibrium, the required time and NO_X concentration are discussed under various reaction conditions. Assumed the change rate of NO_X concentration is less than 0.005, the reaction reached equilibrium. As shown in Fig.2, under 10bar condition, the change rate of NO_X concentration with time is obtained by differential

calculus of this change. When the change rate of NO_X concentration with the time is less than 0.005, we consider that the reaction has reached equilibrium. The time and NO_X concentration are the equilibrium time and concentration of the reaction.

By keeping the same simulation condition and increasing the constant pressure reaction time of the simulation process, the relationship between NO_X concentration and time is analyzed. Using the analysis method of Fig.2, effects of absorption pressure, absorption temperature, initial oxygen concentration and initial NO concentration on equilibrium time and concentration are analyzed systematically.

Effect of Pressure on No Absorption Equilibrium Time and Concentration

The reactor temperature is 25°C, oxygen concentration is 5% and initial NO concentration is 836ppm in mixture 2. By changing the stopping pressure in Rbatch2, and increasing the constant pressure reaction time, effect of pressure on NO absorption equilibrium time and concentration is studied. The pressure range is 4bar to 30bar, and results of the simulation are shown in Fig.3.

As shown in Fig.3, the NO_X equilibrium concentration and time decrease gradually with the increase of pressure. When the pressure is low, the concentration and time of NO_X decrease obviously. When the pressure is high, they reduce gradually. It is obviously that increase of pressure can reduce the NO_X absorption equilibrium time. But when the pressure is too high, it will increase the compression work of denitration process and investment in equipment (especially the acid corrosion).

Effect of Temperature on No Absorption Equilibrium Time and Concentration

The stopping pressure in Rbatch2 is 10*bar*, oxygen concentration is 5%, and initial NO concentration is 836*ppm* in mixture 2. By changing the absorption reactor temperature and increasing the reaction time of absorption process, effect of temperature on NO absorption equilibrium time and concentration is studied. The range of temperature is 10°C to 85°C, and results of the simulation are shown in Fig.4.
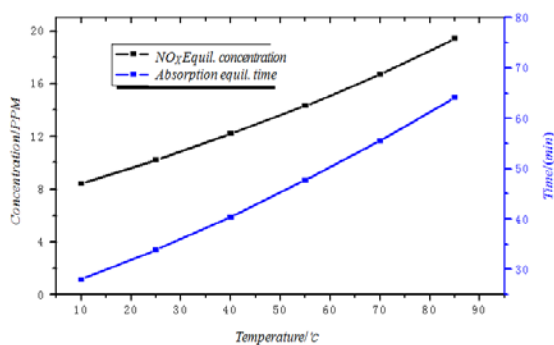

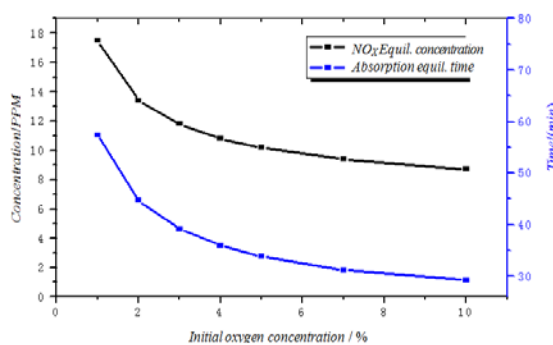Fig.4 Effect of temperature on NO absorption on NO equilibrium time and concentration

Fig.5 Effect of initial oxygen concentration absorption equilibrium time and concentration

As shown in Fig.4, the NO_X equilibrium concentration and time increase almost linearly with the increase of temperature. When the temperature is 10°C, the absorption equilibrium concentration is 8.4*ppm*, and the absorption equilibrium time is 1681*S*. When the temperature increases to 85°C, the absorption equilibrium concentration is 19.4*ppm*, and the absorption equilibrium time is 3852*S*. It shows that increase of temperature has a great effect on the NO_X equilibrium concentration and time. It is unfavorable to reduce NO_X equilibrium concentration and time. But increase of temperature can reduce the energy consumption of flue gas condensation.

Effect of Initial Oxygen Concentration on NO Absorption Equilibrium Time and Concentration

The stopping pressure in Rbatch2 is 10*bar*, reactor temperature is 25°C, and initial NO concentration is 836*ppm* in mixture 2. By changing the oxygen concentration in mixture 2 and increasing the

reaction time of absorption process, effect of initial oxygen concentration on NO absorption equilibrium time and concentration is studied. Initial oxygen concentration range is 1% to 10%, and results of the simulation are shown in Fig.5.

As shown in Fig.5, the NO_X equilibrium concentration and time decrease gradually with the increase of initial oxygen concentration. It shows that increase of initial oxygen content can reduce NO_X equilibrium concentration and time. When the initial oxygen content is 1% to 3%, oxygen content has a great effect on NO_X equilibrium concentration and time. When the initial oxygen content is greater than 5%, the increase of it has little effect. Therefore, the 3%-5% initial oxygen is more suitable for the denitrification process.

Effect of Initial NO Concentration on NO Absorption Equilibrium Time and Concentration

The stopping pressure in Rbatch2 is 10*bar*, oxygen concentration in mixture 2 is 5%, and reactor temperature is 25°C. By changing the NO concentration in mixture 2 and increasing the reaction time of absorption process, effect of initial oxygen concentration on NO absorption equilibrium time and concentration is studied. Concentration range of NO concentration 165*ppm* to 950*ppm*, and results of the simulation are shown in Fig.6.
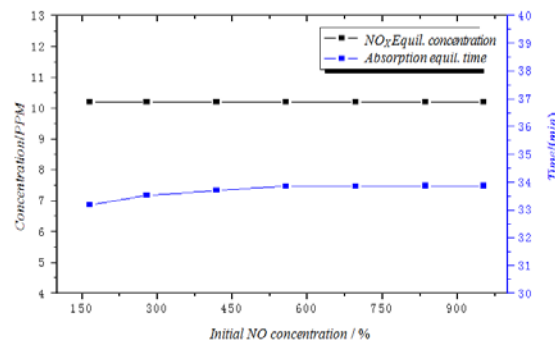

Fig.6 Effect of initial NO concentration on NO absorption equilibrium time and concentration

As shown in Fig.6, the NO_X equilibrium concentration is almost unchanged with the increase of initial NO concentration. The NO_X equilibration time increases slightly and it happens almost at low initial NO concentration. During the initial NO concentration change from 165*ppm* to 950*ppm*, the equilibrium time of absorption increases only 40*S*. Thus, increase of initial NO concentration has almost no effect on the NO_X equilibrium time and concentration. Although oxy-fuel combustion can significantly reduce NO_X emissions, but because of the flue gas recirculation, the NO_X concentration remains high in flue gas even higher than that under air combustion. Therefore, the pressure denitrification process is more suitable for oxy-fuel combustion.

Optimization Analysis of Pressure Denitrification Process

According to above balance calculation, oxygen concentration range of oxy-fuel combustion process is 3% to 5%. Selected the median value of 4%, the content of NO_X in the flue gas had little effect on the pressure denitrification process, so the selection of it has no effect. Therefore, the content of NO is set as 850*ppm*, a typical one. The lower denitrification temperature is more beneficial to the denitrification process of pressurized flue gas. But the temperature is too low, the energy consumption and investment in equipment will increase. Based on the above factors, the room temperature of 30°C is more appropriate. Therefore, the choice of pressure during pressurization is critical[8,9].

The pressure of flue gas should be increased before pressure denitrification. If centrifugal compressor is used to compress the flue gas, the power consumption of the compression process is related to exhaust capacity, ambient temperature, pressure ratio, efficiency, and so on. The formula is as follows[10],

$$W = \frac{\rho V_K RT \ln(P_2 / P_1)}{3600\eta_T\eta_M}$$

(1)

Among: ρ, density of flue gas under standard conditions, kg / m^3; V_K, exhaust capacity of air compressor, m^3 / h; R, gas constant, $R = 0.278 kJ / (kg \cdot K)$; T, ambient temperature, K; P_2, exhaust pressure, Mpa; P_1, intake pressure, Mpa; η_T, isothermal efficiency of air compressor; η_M, mechanical efficiency.

Because the intake pressure of air compressor is 0.1Mpa, the compression power consumption is proportional to W, that is $W=N\ln P_2$ (N is a constant constant). The ratio of compression power consumption increment of 2bar pressure increased and the amount of NO_X removal is the power consumption of unit NO_X removal. This value is used as the ordinate, and the pressure is the abscissa. The curve is drawn as shown in Fig.7.
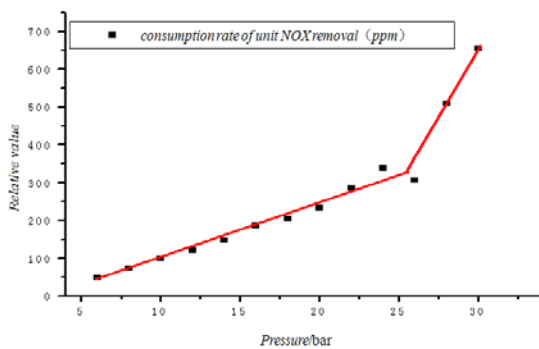

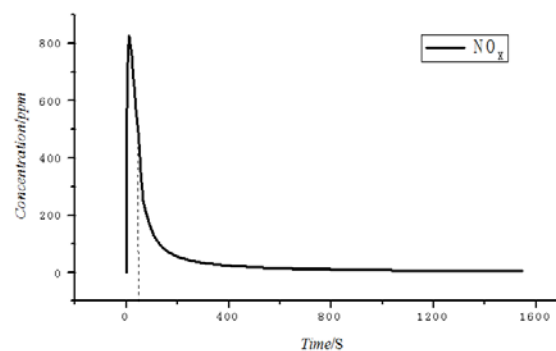Fig.7 Relationship between pressure and unit power consumption of NO_X removal

Fig.8 Relationship between NO_X concentration and time in optimized conditions

As shown in Fig.7, the power consumption rate of NO_X increases gradually with the increase of pressure. But when the pressure is below 26bar, the consumption rate of unit NO_X removal increases slowly with the increase of pressure. When the pressure is higher than 26bar, it increases rapidly. So we choose 26bar as the reaction pressure for the pressure denitrification process. Finally, the Aspen Plus process simulation is established according to the optimization conditions. The relationship between the concentration of NO_X and time is shown in Fig.8.

Simulation results show that NO_X declines to a great extent at the end of pressure boosting. However, the removal rate of NO_X is only 43.9%. When the residence time is 147S, the removal rate of NO_X can reach 90%. When the residence time is 249S, the removal rate of NO_X can reach 95%. When the residence time is 1363S, the equilibrium concentration is 6.5ppm.

Summary

With the increase of pressure, initial oxygen content and the decrease of temperature , the equilibrium concentration and time of pressure denitrification decrease gradually. The content of NO in flue gas has little effect on equilibrium concentration and time. Finally, according to analysis and calculation, the temperature of 30° C and the pressure of 26bar are more suitable for pressurized flue gas denitrification. Finally, the optimized working condition is obtained under the condition of initial oxygen content of 4% and initial NO concentration of 850ppm. It indicates that the removal rate of NO_X is only 43.9% at the end of pressure boosting.When the residence time is 147S, the removal rate of NO_X can reach 90%. When the residence time is 249S, the removal rate of NO_X can reach 95%. When the residence time is 1363S, the equilibrium concentration is 6.5ppm.

Acknowledgement

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