Optimization and control of NOx in coke oven vertical flue under flue gas recirculation

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Abstract. The orthogonal experimental design with CFD simulation was carried out to simulate combustion in the coke oven vertical flue at external flue gas recirculation. The control of nitrogen oxides (NOx) generation was analysed by different factors such as air excess coefficient, oxidant preheating temperature, flue gas recirculation and oxidizer inlet cross-sectional area ratio. The simulation results showed that the the orthogonal experimental design with CFD had a significant effect on the combustion process. The flue gas recirculation ratio effectively improved the temperature distribution uniformity in the vertical flue, and had a significant effect on NOx production in the vertical flue. Within the parameters of the orthogonal design, when the air excess coefficient was 1.1, the oxidant preheating temperature was 1200 K, the flue gas recirculation ratio was 30%, and the oxidant inlet cross-sectional area ratio was 0.5, the NOx production in the coke oven vertical flue was the smallest, lower than the industry emission standard.

1. Introduction

Coking industry is one of the most important supporting industries of steel industry. Coke acts as the reducing agent and heat source in the reduction process of iron ore. However, the production process of coke also generates a series of pollution problems, such as soot, NOx, SO2, etc. Among them, NOx is mainly produced by the combustion process of coke oven gas (blast furnace gas) in the coke oven combustion chamber. Currently, reducing NOx in flue gas is mainly divided into the two methods: source control and terminal control. The source control is to inhibit the formation of NOx from the root. It mainly includes staging combustion technology, thick and thin combustion method, flue gas recirculation technology and so on. Poraj et al. [1] found that air staging combustion technology had an active role in coke oven heating system. Smolka et al. [2] conducted two-dimensional simulation of coke oven vertical flue under the condition of smoke external circulation by controlling the mass flow of oxidant inlet constant, and concluded that when smoke external circulation ratio was 0.2-0.25, the production amount of NOx was the smallest. Terminal treatment of reducing NOx in flue gas includes biological method, physical method and chemical method, etc.[3]. Mature technologies on the market mainly includes SCR, SNCR and dual ammonia solution[4].

In this paper, numerical simulation was carried out for the single opposing vertical flue in the combustion chamber of JN60 coke oven which are widely applied in coking industry, and the external flue gas circulation and the combustion process in the coke oven vertical flue are considered as a whole, which is regarded as a strengthening effect of flue gas internal circulation. Through the
orthogonal design method, the simulation calculation of the combustion process under the external circulation of flue gas in the vertical flue of coke oven was carried out.

2. Model and calculation method

2.1. Physical model
Figure 1 shows the three-dimensional structure diagram and the two-dimensional top view of the coke oven vertical flue. The coke oven gas occurs in the combustion process in the upward vertical flue, and then flue gas produced passes through the bridge window into the downward vertical flue. Most of the flue gas discharge through outlet while the remaining through recirculation window participates in combustion process with air and coke oven gas [3]. The model of coke oven vertical flue is divided into structured hexahedral grids with a total number of 157,780 grids which shows good grid quality.

![Figure 1. The three-dimensional structure diagram (left) and the two-dimensional top view (right) of the coke oven vertical flue.](image)

2.2. Mathematical model
The turbulence model was the standard k-ε model. The radiation model was the P-1 model. The combustion model was modelled by material transport and eddy-dissipation model was analysed the turbulence-chemistry interaction. The NOx model was the thermodynamic NOx model.

2.3. Boundary conditions of model
Coke oven vertical flue is mainly composed of solid furnace wall and gases which join combustion reaction. The furnace wall of coke oven vertical flue is composed of silicon bricks whose density is 2323 kg/m³ and specific heat capacity is 858 J/(kg·K). The average thermal conductivity of that is 1.90 W/(m·K). The density of coke oven gas is approximately 0.45 kg/m³. As a secondary energy source, coke oven gas is complex in composition. In this study, a quantitative standard was adopted to conduct numerical simulation. The coke oven gas is composed of 12% H₂, 41% CH₄, 19% CO, 19% N₂ and 9% CO₂ (mass fraction), and air is composed of 23% oxygen and 77% nitrogen (mass fraction).

2.4. Circulation model of flue gas
In order to simulate the flue gas recirculation combustion process in the coke oven vertical flue, the combustion in the coke oven vertical flue and the flue gas circulation are regarded as a system. In this case, the flue gas external circulation is actually an intensification method for the flue gas internal circulation process, as shown in the figure 2:

Formulas involved in the circulation of flue gas in coke oven vertical flue:

\[
\begin{align*}
  m_{OX} &= m_A + R \cdot m_{EX} \quad (1) \\
  R &= m_R/m_{EX} \quad (2) \\
  y_{OX} &= (y_{EX} \cdot m_R + y_{AI} \cdot m_A)/m_{OX} \quad (3)
\end{align*}
\]
Where $m_{\text{OX}}$, $m_{\text{EX}}$ and $m_{\text{F}}$ respectively represent the mass flow rate of oxidant, mass flow rate of air and the mass flow rate of coke oven gas. $m_{\text{R}}$ is the flue gas recirculation mass flow rate. $y_{\text{OXi}}$, $y_{\text{EXi}}$ and $y_{\text{Ai}}$ respectively are the mass fraction of component $i$.

![Figure 2. The combustion system of coke oven vertical flue under the flue gas circulation.](image)

3. Orthogonal experimental design

The air excess coefficient ($\alpha$), the cross-sectional area ratio of the oxidant inlet ($S_r$), the pre-heating temperature of the oxidant inlet ($T_{\text{OX}}$) and the flue gas recirculation ratio ($R$) of the coke oven vertical flue were taken as the main influencing factors on the combustion process in the coke oven vertical flue. The table 1 was designed for numerical calculation of the vertical combustion process. And each factor was divided into four levels. And the $L_{16}(4^5)$ orthogonal table of the vertical flue combustion process was designed.

| Factors      | levels |
|--------------|--------|
| $\alpha$     | A 1.05 1.10 1.15 1.2   |
| $T_{\text{OX}}$ (K) | B 1000 1300 1100 1200 |
| $R$ (%)      | C 10 30 20 0     |
| $S_r$        | D 1 0.25 0.5 0.75 |

4. Results and discussion

4.1. Orthogonal design-direct analysis

Orthogonal design-direct analysis of the experimental results was shown in table 2 and 3. $K_i$ ($i =1,2,3,4$) represents the sum of experimental results with the same level number in any column. And $k_i$ is the arithmetic mean value of the experimental results with the same level number in any column. $R$ represents the range and $R= \text{Max} \{k_1, k_2, k_3, k_4\} - \text{min} \{k_1, k_2, k_3, k_4\}$ in any column. The range is positively correlated with the experimental results. Therefore, according to the intuitive analysis of experimental results, the priority order of factors affecting experimental results was CDBA. The range of factor C was the most influential, and in other word, the main factor affecting the generation of NOx was the flue gas recirculation ratio. At the same time, in order to reduce the NOx generation in the combustion process of coke oven vertical flue, the minimum value of any column of $K_i$ should be selected as the evaluation index of the optimal scheme. After comparison, the optimal scheme was $A_2B_2C_3D_3$. As shown in table 4, The experimental results of the optimal scheme and the orthogonal experimental group were in comparison. It could be found that both number 8 and optimal theme can...
effectively reduce NOx generation in coke oven vertical flue. The optimal scheme had the best effect and belongs to the optimal scheme.

Table 2. Orthogonal design-direct analysis of the experimental results

| number | A  | B  | Error | C  | D  | y(NOx) | NOx(mg/m³) | Tmean(K) | Tmax(K) |
|--------|----|----|-------|----|----|--------|------------|----------|---------|
| 1      | 1  | 1  | 1     | 1  | 1  | 0.00369| 826        | 1781     | 2430    |
| 2      | 1  | 2  | 2     | 2  | 2  | 0.00216| 523        | 1749     | 2402    |
| 3      | 1  | 3  | 3     | 3  | 3  | 0.00084| 215        | 1676     | 2255    |
| 4      | 1  | 4  | 4     | 4  | 4  | 0.00554| 1304       | 1824     | 2501    |
| 5      | 2  | 1  | 2     | 3  | 4  | 0.00181| 443        | 1734     | 2355    |
| 6      | 2  | 2  | 1     | 4  | 3  | 0.00491| 1151       | 1759     | 2466    |
| 7      | 2  | 3  | 4     | 1  | 2  | 0.00492| 1155       | 1820     | 2517    |
| 8      | 2  | 4  | 3     | 2  | 1  | 0.00018| 50         | 1555     | 2170    |
| 9      | 3  | 1  | 3     | 4  | 2  | 0.00562| 1298       | 1846     | 2562    |
| 10     | 3  | 2  | 4     | 3  | 1  | 0.00557| 1190       | 1772     | 2559    |
| 11     | 3  | 3  | 1     | 2  | 4  | 0.00106| 269        | 1663     | 2337    |
| 12     | 3  | 4  | 2     | 1  | 3  | 0.00022| 60         | 1604     | 2155    |
| 13     | 4  | 1  | 4     | 2  | 3  | 0.00099| 226        | 1663     | 2360    |
| 14     | 4  | 2  | 3     | 1  | 4  | 0.00525| 1098       | 1839     | 2573    |
| 15     | 4  | 3  | 2     | 4  | 1  | 0.00526| 1162       | 1756     | 2547    |
| 16     | 4  | 4  | 1     | 3  | 2  | 0.00436| 898        | 1840     | 2584    |

Table 3. Orthogonal design-direct analysis of the NOx generation

| factor | A   | B   | Error | C   | D   | NOx(mg/m³) |
|--------|-----|-----|-------|-----|-----|------------|
|        | K1  | K2  | K3    | K4  | k1  | k2  | k3  | k4  | R   | A2  | B3  | C2  | D3  |
|        | 2869| 2798| 2816  | 3383| 717 | 700 | 704 | 846 | 146 | 2   | 3   | 4   | 1   |
|        | 2793| 3962| 2801  | 2312| 698 | 990 | 700 | 578 | 413 | 4   | 2   | 3   | 4   |
|        | 3143| 2189| 2661  | 3874| 786 | 547 | 665 | 968 | 421 | 3   | 3   | 4   | 1   |
|        | 3138| 1067| 2746  | 4915| 785 | 267 | 686 | 1229| 962 | 2   | 3   | 4   | 1   |
|        | 3227| 3874| 1652  | 3114| 807 | 968 | 413 | 779 | 555 | 3   | 3   | 4   | 1   |

According to the simulation results of orthogonal design experiment, the horizontal trend chart of each factor was drawn. As shown in figure 3 (a), (b), (c), (d), with the increase of air excess coefficient and oxidant pre-heating temperature, the formation of NOx did not change significantly, while the ratio of flue gas external circulation ratio and the cross-sectional area of oxidant inlet had an obvious influence on the formation of NOx. From the orthogonal test table, it could be found that when the ratio of flue gas external circulation was 30% and the cross-sectional area ratio of oxidant inlet was 0.5, the production amount of NOx was the smallest.

4.2. Analysis of variance

The intuitive analysis method of orthogonal design is simple and intuitive. However, the intuitive analysis method cannot estimate the magnitude of the error, and cannot accurately estimate the importance of each factor to the experiment results. Therefore, analysis of variance is used to further analyse the orthogonal test group.
As shown in table 5, after calculating the deviation sum of squares (SS), degrees of freedom (df) and mean square of the experimental results of the orthogonal design group, it could be found that the factor MS_A<MS_E, which indicated that factor A was a secondary factor to the generation of NOx in the vertical flue. Therefore, factor A could be put into the Error. Then, the deviation sum of squares, degrees of freedom and mean square of Error all changed.

Table 5. Sum of squares of the experimental results

| Factors | SS        | df | MS       |
|---------|-----------|----|----------|
| A       | 2.60E-06  | 3  | 8.68E-07 |
| B       | 8.20E-06  | 3  | 2.73E-06 |

According to the F distribution table, F_{0.05}(3,6) = 4.76, in which F_C>F_{0.05}(3,6), so factor C had significant influence on the experimental results. While F_B<F_{0.05}(3,6) and F_D<F_{0.05}(3,6), which indicated that factor B and D had no significant influence on the experimental results. Therefore, it could be found that the generation of NOx in the vertical flue of coke oven was mainly affected by the recirculation ratio of flue gas. It was because the flue gas recycling effectively reduced the partial pressure of O_2 in the air under the condition of a certain oxygen content, the maximum temperature in the vertical flue decreased significantly.
Table 6. New sum of squares of the experimental results

| factors | SS    | df | MS    | F      | Significance |
|---------|-------|----|-------|--------|--------------|
| B       | 8.20E-06 | 3  | 2.73E-06 | 1.578  | *            |
| C       | 3.62E-05 | 3  | 1.21E-05 | 6.994  | *            |
| D       | 1.41E-05 | 3  | 4.69E-06 | 2.711  | *            |
| $e^\Delta$ | 1.04E-05 | 6  | 1.73E-06 |         | *            |
| SUM     | 6.11E-05 | 15 | 4.07E-06 |         | *            |

5. Conclusions
In order to better analyse the generation of NOx in the vertical flue of coke oven under flue gas recirculation, the orthogonal design and numerical simulation method were adopted, and remarkable results were obtained. In the case that the heat flux density was guaranteed, the orthogonal test results showed that when the air excess coefficient was 1.1, the oxidant preheating temperature was 1200K, the flue gas recirculation ratio was 30%, and the oxidant inlet cross-sectional area ratio was 0.5, the NOx generation in the coke oven vertical flue was the minimum, which was 44mg/m$^3$. There was no doubt that the environmental protection requirements of coking industry production. The flue gas recirculation had a significant effect on the formation of NOx in the vertical flue. While the air excess coefficient, the preheating temperature of the oxidant and the proportion of the inlet cross-sectional area of the oxidant have no obvious influence on the formation of NOx.

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