Modeling and Feedforwad Control of Flue Gas Denitration in Coking Reversing Process

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***Abstract*—There is a large amount of energy waste in coking flue gas desulfurization and denitrification integrated device during the revising process of coking, in order to solve this problem, aiming at the unique reversing process of coking, the reasons for the change of NO_x concentration in this process are deeply analyzed; the disturbance of inlet and outlet NO_x concentration during the reversing process is modeled by a proposed data driven identification method, the verification is carried out based on the actual data of two 55-hole and 6-meter top charging coke ovens, according to the statistical data, the relationship model of inlet and outlet NO_x concentration loss is obtained; we designed a feedforward control system and the feed forward control rate of O_3 is calculated based on the outlet disturbance model, the power consumption cost savings is obtained through simulation, which indicates the necessity of the work in this paper.**

Keywords—process control; dynamic modeling; coking; revising process; denitration

I. INTRODUCTION

China is the world's largest coking production country[1], with a huge annual emission of sulfur dioxide and nitrogen oxides. With the formal implementation of "coking chemical pollutant emission standards" in January 1, 2015, the sulfur dioxide and nitrogen oxides emission targets of coking industry are put forward strict and clear quantitative requirements[2]. While the disposal of flue gas sulfur dioxide and nitrogen oxide in the coking process of domestic coking industry, is almost in a blank state: flue gas is directly released into atmosphere through the chimney. Under this situation, Jiangxi Coking & Chemical Co. Ltd. in the domestic has taken the lead in building and operating of coking gas desulfurization and denitrification integrated engineering, which adopts process flow of wet ammonia process super turbulent desulfurization and two sections of forced oxidation of urea denitrification[3].

The specific process is: Flue gas from coking process is fed into waste heat recovery boiler through draught fan, and the temperature is reduced from 300℃ to 160℃, after the booster fan, the flue gas converges with ozone from the ozone input pipe before entering the desulfurization tower, part of nitric oxide in flue gas can rapidly react with ozone, converting into nitrogen dioxide. Then flue gas enters the concentrated section of desulfurization tower, after spraying and washing, cooling to 60 ℃ , then the gas enters the absorption section of desulfurization tower through the gas cap, countercurrent contacting with desulfurization absorption liquid sprayed from the top of desulfurization tower, sulfur dioxide in flue gas react with ammonium sulfite in the absorption liquid, converting into ammonium hydrogen sulfite. As a result, sulfur dioxide is removed and purified, in the process, the urea residue comes from the bottom of the denitrification tower is intermittently injected in order to realize pre denitrification. The liquid at the bottom of the absorption section backflow to the liquid storage section and oxidation section at the bottom of desulfurization tower, in order to restore the absorption capacity of the absorption liquid, ammonia has to be supplemented. Process water is sprayed from the top of desulfurization tower to keep the liquid level of the desulfurization tower in a reasonable range. Air is drummed into the liquid storage tank, part of the ammonium sulfite in the liquid storage tank is oxidized to ammonium sulfate in order for the spray, evaporation and concentration in the concentration section and other subsequent processing.

The flue gas pipeline after desulfurization is connected to ozone input pipe, ozone is mixed into the flue gas under the temperature about 60℃, part of nitric oxide in flue gas can rapidly react with ozone, converting into nitrogen dioxide, countercurrent contacting with desulfurization absorption liquid sprayed from the top of denitrification tower after entering the lower part of the tower, the reduction reaction of nitric oxide and nitrogen dioxide in the flue gas urea in solution generating nitrogen, carbon dioxide and water, the denitrification process is completed. The flue gas reaches the environmental protection emission standard is discharged into the atmosphere at the top of denitrification tower, and the whole process of the flue gas treatment is completed.

The integration process device of desulfurization and denitrification as shown in figure 1.

After analysis, the main factors affecting the effect of denitrification are ozone input, urea solution (nitrate absorption liquid) concentration and circulation[4-8]. At present, the process of denitrification mainly rely on manual control, and due to the urea solution concentration cannot be measured, the current control method is to maintain the urea cycle, the ozone generator power determined by operator according to the average NO_x concentration of the tower inlet and outlet.

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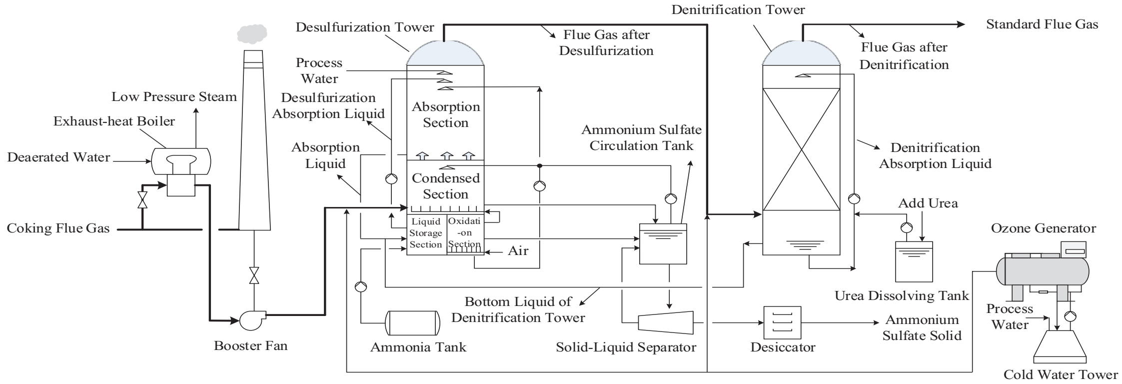


Fig.1 Desulfurization and denitration integrated device for coking flue gas

Because of A. the existence of "reversing" operation in coke oven, there is a large range of fluctuations of flue gas composition during reversing period[9](Shown in Fig. 3); B. In the process of denitrification, the cost of the ozone generator accounts for more than 80% of the total cost of the whole device[10]. So, if the production of ozone in the revising process is not controlled but make it still running at the power of non-revising-process, will lead to a huge waste of energy, greatly increasing the cost of enterprises, which is not conducive to the economic operation of the device. In addition, due to C. the flue gas inlet is far from the outlet, there is a large lag in the process of flue gas concentration detection. If we take regular feedback control, it will inevitably cause untimely control, even results in the outlet flue gas concentration exceeds the permissible standard.

In order to solve the above problem, the disturbance modeling of coking flue gas NO_x during revising process and ozone output feedforward control needs to be carried out[11]. The paper combined the revising process and generation mechanism of NO_x to analysis the reasons for the impact of NO_x concentration; we proposed a method of modeling and identifying the NO_x disturbance by using the response curve of continuous pulse signal and first order inertia transfer function, based on the flue gas concentration measured data of two 55-hole and 6-meter top charging coke ovens in the coking plant, the validity of the model is verified by comparison; Feedforwad control system is designed and the feedforward control rate of ozone is obtained based on the invariance principle[12] and simulation. After calculating single day energy saving costs, the results show that the main significance of this paper.

II. ANALYSIS AND MODELING OF NO_X CONCENTRATION DISTURBANCE IN REVERSING PROCESS

A. NO_x Disturbance Analysis

The coking reversing process as shown in Figure 2[13].

We can know from the literature [14] that there is almost entirely thermal-type nitrogen oxides under lean gas heating. The gas for the coking plant is mixed with a small amount of coke oven gas in addition to the lean gas, so the generated NO can be considered all thermal - type nitrogen oxides; the N and O in thermal-type NO are all derived from the air, while there is a great change in the air quantity during the reversing process. In summary, the generation NO_x quantity in the reversing process related to combustion temperature and air quantity.

When reversing process begins, gas closed first and air is excessive, the disappearance of high temperature zone and the increase of air composition cause the NO_x concentration of waste gas dropping significantly, however, NO_x concentration will not drop to 0 due to the fuel-type NO, coke oven leakage and the short period of reversing time and other factors. After about 40 seconds, gas is gradually fed, but the gas is in the original down flue, where is at the lowest temperature before the reversing process, so affected by ambient temperature, the actual combustion temperature of the flame cannot reach the theoretical combustion temperature, as a result, NO_x could not quickly return to normal values until the temperature of flame reaches the theoretical value with the increasing of the temperature of vertical heating flue.

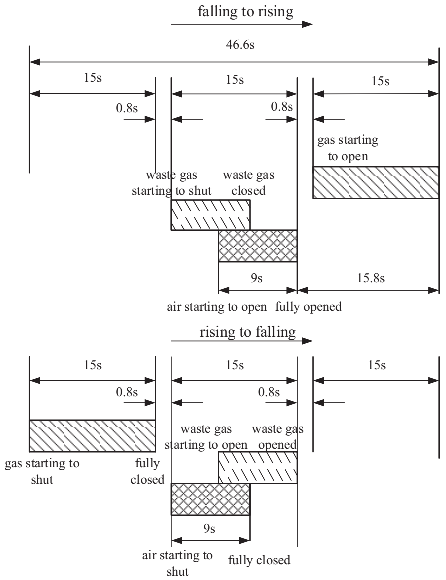


Fig.2 Brace operating diagram during reversing process

Take 1 hour flue gas NO_x concentration data of the coking plant as shown in Fig.3, the correctness of the above analysis can be verified by the two reversing process at 05:15 and 05:45. NO generating quantity is affected when O_2 content is at the lowest point during each reversing process, as shown in the inflexion point in red circle, but it can be ignored because of the minimal impact.

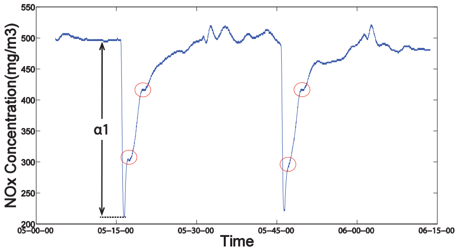


Fig.3 NOx concentration of flue gas in reversing process

B. *Modeling of NO_x Concentration Perturbation*

In this paper, the disturbance of generated NO_x after the flue gas enters the desulfurization and denitrification device, the disturbance degree could be changed due to various factors, called as outlet NO_x disturbance. Obviously, the outlet NO_x disturbance is the object of feedforward control. The paper will model the inlet and outlet NO_x disturbance respectively.

Because the NO_x concentration is affected by the temperature and the oxygen content in the air, while the temperature of heating wall and the oxygen content in the air are extremely complex, and the carbonization chamber leakage will also affects NO_x concentration. As a result, it's quite difficult to model the NO_x disturbance during reversing process relying on the mechanism[15].

Desulfurization and denitrification site has accumulated a large number of process data and information, providing precondition for data driven process modeling method[16]. Based on this, we adopt a method of data driven modeling, the structure of general disturbance model is firstly constructed, the loss relation models of inlet and outlet NO_x concentration under different conditions are obtained by statistics, the parameters of inlet and outlet disturbance model are identified by analytical method.

1) *Modeling of inlet NO_x disturbance*

It is found that the concentration disturbance process is very similar to the response curve of pulse signal in series with one order inertia transfer function, so we put rectangular pulse with cycle P and width W in series with an unit ratio coefficient first order inertial link $\mathrm{G} = \frac{1}{\mathrm{T}*\mathrm{s}+1}$ as the disturbance model.

The following facts are obtained through statistical analysis of the measured data:

- The reversing process occurs once every 30 minutes, that is, the disturbance period is $1800/\tau$ (τ is the DCS sampling period, 1 second).
- It takes about 40 seconds of flue gas concentration from starting to drop to the lowest point, and about 800 seconds from the minimum to stationary state.

At this point, we can first determine 2 unknown coefficients P and W as 1800 and 40 respectively. The response curve is shown in Figure 5(a).

Divide the working conditions of the coking process according to the factors influencing inlet flue gas NO_x concentration to S={S_1, S_2……S_n}, define the steady inlet NO_x concentration corresponding to above conditions as C={C_1, C_2……C_n}, define the minimum NO_x concentration corresponding to above conditions during reversing process as C'={C_1', C_2'……C_n'}. Here, n denotes the number of working conditions, $n \in N$.

Define the loss of inlet flue gas NO_x concentration in reversing process as $\alpha_1 = f_1(C) = \mathrm{C} - \mathrm{C}', (\alpha > 0)$, representing the degree of the impact of reversing process on NO_x with different concentration(Figure 3).

The relationship between α_1 and C is obtained based on the historical data(Figure 4).

In order to improve the fitting precision, take the mean value of the horizontal coordinate as the center, and the standard deviation as the normalization factor[17], obtained coefficient matrix P=[36.81 -43.21 142.7 439.7] via identifying, the cubic function loss model is

$$f_1(C) = p(1) * C^3 + p(2) * C^2 + p(3) * C + p(4) \quad (1)$$

Various indicators as shown in Table 1.

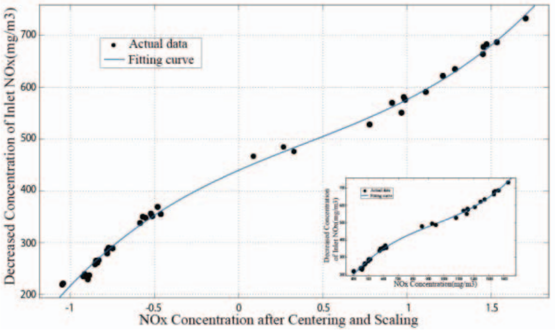


Fig.4 Relationship between NO_x concentration and the loss of inlet NO_x in reversing process

TABLE I. MODELING PERFORMANCE INDEX

Index	Value
SSE	3191
R-square	0.9968
RMSE	10.31

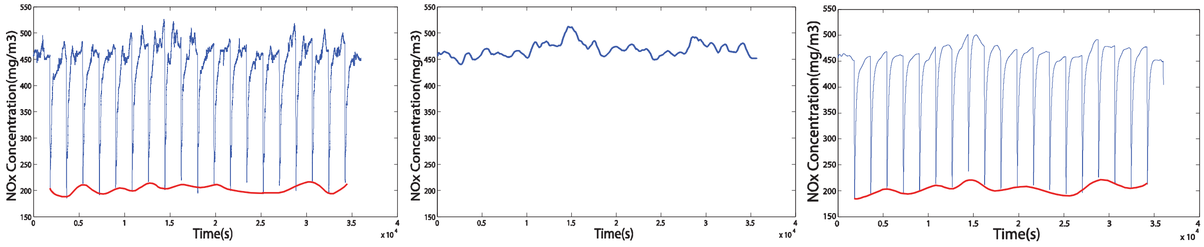
The steps of identifying the unknown parameters h and T using analytic method are as follows:

Determine the class n of current working condition, so we can get the average NO_x concentration C_n, then the loss extent A can be calculated by model(1).

The time domain response formula of the concentration disturbance model is

$$\mathrm{y} = \begin{cases} h\left(1 - \exp\left(-\frac{t}{T}\right)\right) & 0 \le t \le 40 \\ h\left(1 - \exp\left(-\frac{t}{T}\right)\right) - h\left(1 - \exp\left(-\frac{t-x}{T}\right)\right) & 40 \le t \end{cases} \quad (2)$$

We can get the following equations:



(a) (b) (c)

Fig.6 Verification of NO_x disturbance modeling effect during reversing process

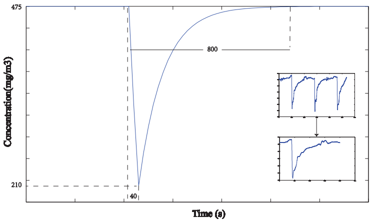
$$
\begin{cases}
h\left(1-\exp\left(-\dfrac{40}{T}\right)\right)=C_n{}'-C_n=-\alpha_1 \\
C_n+h\left(1-\exp\left(-\dfrac{t}{T}\right)\right)-h\left(1-\exp\left(-\dfrac{t-40}{T}\right)\right)+\varepsilon=C_n \\
\text{Here } \mathrm{t}=800-40=760
\end{cases}
\quad (3)
$$

Where $\varepsilon \in \mathbb{R}^+$, usually take (0,10], the purpose is to remain a certain margin to avoid the response reaching the steady-state value prematurely. Take 0.5 as the step size, the least square method is used to obtain the optimal solution of the above nonlinear implicit equation Where $\varepsilon \in (0,10]$, so the parameter h and T can be identified.

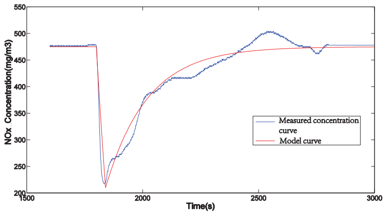
2) Validation of inlet NO_x disturbance model

Under current coking conditions, C=475 mg·m^{-3}, according to the loss model, the loss concentration can be determined as α_1=265. After calculation, the curve fitting degree is better when $\varepsilon = 5$, at this time h=-1330, T=180. Compared with the actual concentration curve of a certain reversing process. As shown in Figure 5(b), the effect is satisfactory.

Choose any 10 hours NO_x concentration data,(Figure 6(a)), set a time window for 900 seconds, the data in the window is processing by interpolation according to the data of the first time and the last time. Execute once every 1800 seconds to remove the disturbance of reversing process. Then the moving average filter is used to smooth the data processing[18], removing the data noise interference, as shown in Figure 6(B). Add the established disturbance model in the concentration data after removing the disturbance and smoothing, as shown in Figure 6(C). Compared with Figure 6(a), the trend and amplitude of the disturbance are consistent with the measured data, which proves the validity of the proposed model.



(a)



(b)

Fig.5 Inlet NO_x perturbation curve during reversing process and comparison

3) Modeling of outlet NO_x disturbance

Let steady state outlet NO_x concentration D={$\mathrm{D1}(\Theta,\varepsilon)$, $\mathrm{D2}(\Theta,\varepsilon)$……$\mathrm{Dn}(\Theta,\varepsilon)$} corresponding to different working conditions S, and the corresponding mean minimum NO_x concentration D’={$\mathrm{D1}(\Theta,\varepsilon)'$， $\mathrm{D2}(\Theta,\varepsilon)'$…… $\mathrm{Dn}(\Theta,\varepsilon)'$} in reversing process.

Here, n denotes the number of working conditions, $n \in N$; $\Theta(\mathrm{C}_p, \mathrm{v}, \mathrm{O}_3, \mathrm{L})$ denotes the stable operating point of the desulfurization and denitrification device: C_p is inlet NO_x concentration of the desulfurization tower under working condition S_p, V is inlet flue gas flow rate, O_3 is total input of two stage ozone, L is the cycle solution of urea in denitrification tower; ε stands for uncontrollable and uncontrollable factors such as urea solution concentration, sensor error and environmental factors etc.

Defined the loss of outlet flue gas NO_x concentration in reversing process as $\alpha_2 = f_2(\Theta,\ \varepsilon) = \mathrm{D}(\Theta,\ \varepsilon) - \mathrm{D}(\Theta,\ \varepsilon)'$, $(\alpha > 0)$, representing the degree of the impact of reversing process on NO_x with different concentration.

$\mathrm{C}_p$and O_3in Θ are the most significant variables for α_2, and $\mathrm{C}_p$is related to working conditions of coking process as well as controllable. The relation of $\alpha_2 - (\mathrm{C}_p, \mathrm{O}_3)$ as shown in Figure 7.

In the graph, the curves of different colors represent different working conditions (S/C). Δp is the difference of outlet flue gas NO_x concentration between the steady state and the lowest point in working condition Sp and $\Theta(\mathrm{C}_p, \mathrm{v}, \mathrm{O}_3 = 0, \gamma)$; Δ is mainly determined by NO_x concentration of the flue gas from the desulfurization tower, which is similar to Figure 4 and can be expressed as Δ= $f_1{}'(C)$.

After the working condition is determined, α_2 has a direct correlation with O_3, and it decreases with O_3 increasing. But when O_3 is the critical value m, it can be considered that the oxidation rate of NO in flue gas reaches the limit[2], so the effect of O_3 on α_2 is minimal.

Compared with inlet flue gas, the structure, P and W of the disturbance model in reversing process are the same as the inlet model except for the loss of concentration.

The identification equation is as follows:

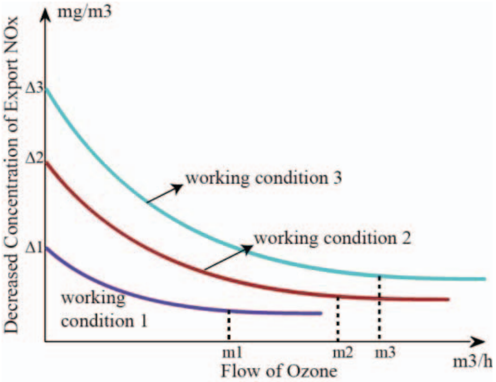


Fig.7 Relationship between the loss of outlet NO_x concentration in reversing process and flow of O_3

$$\begin{cases} h\left(1-\exp\left(-\frac{40}{T}\right)\right)=D_n'-D_n=-\alpha_2=f_2(\Theta,\ \varepsilon) \\ D_n+h\left(1-\exp\left(-\frac{t}{T}\right)\right)-h\left(1-\exp\left(-\frac{t-40}{T}\right)\right)+\varepsilon=D_n \\ \text{Here } \mathrm{t}=800-40=760 \end{cases} \tag{4}$$

4) Validation of outlet NO_x disturbance model

Choose data of the same working condition as section 1.2.2, that is, C=475 mg·m^{-3}, v=10.8-11.3 m·s^{-1}, O_3=1050m^3·h^{-1}, L=150 m^3·h^{-1}.

After calculation, we found the curve fitting degree is good when $\varepsilon=0.5$, and h=-530.331, T=127.299. Compared with the actual concentration curve of a certain reversing process, the effect is satisfactory (Figure 8).

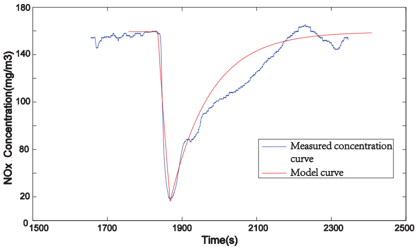


Fig.8 Outlet NO_x perturbation curve during reversing process and comparison

III. FEEDFORWARD CONTROL OF OZONE IN REVERSING PROCESS

A. Control Strategy

In order to simplify the calculation, the amount of ozone is total amount of two towers, the adjustment process is mainly based on single tower, the control channel model is approximate to linear relation.

A typical open-loop dynamic feedforward control system is used[19-20] (Figure 9), where FC is feed forward controller, O_{31} and O_{32} is the ozone enter the desulfurization tower respectively, other letters are the same meaning as section 1.2.3. The feedforward controller determines NO_x disturbance model in reversing process according to the current flue gas NO_x concentration the total amount of ozone and the crculation quantity mount of urea so as to determine the amount of ozone compensation.

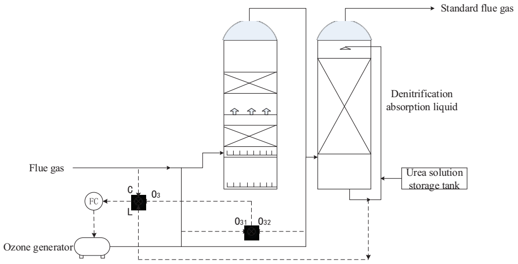


Fig.9 Feedforward control strategy

The transfer function is described as shown in Figure 10.

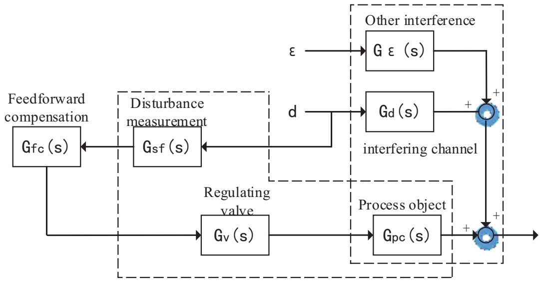


Fig.10 Transfer function description of feedforward control system

Incorporating the sensor and the regulator characteristics into the object (Gpc (s)), the feedforward controller transfer function is obtained according to the invariance principle:

$$\mathrm{Gfc(s)}=-\frac{Gd(s)}{Gpc(s)} \tag{5}$$

B. Simulation and Calculation

We get the disturbance channel transfer function Gd = $\frac{1}{127.299S+1}$ by section 1.2.4, the control channel model is estimated as Gpc = $\frac{-6.3}{7S+1}$ according to the working condition of section 1.2.2, so the feedforward controller Gfc = $-\frac{Gpd}{Gpc}=\frac{7S+1}{801.9837S+6.3}$. Simulation is realized in Simulink(Figure 11).

Due to the cycle of the reversing process is fixed, the feedforward control can be used in a timing mode, so the lead and lag of time can all be realized. In order to simplify the calculation, the pure lag link is omitted in the simulation process.

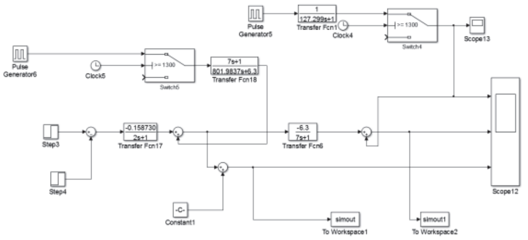
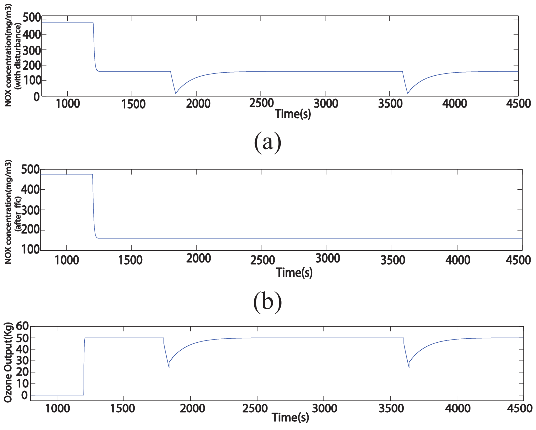


Fig.11 Feedforward control simulation

The control effect is as follows.



(a)

(b)

(c)

Fig.12 Feedforward control effect during reversing process

(a) is the concentration change of NO_x in outlet of the denitrification tower without feedforward control, it can be seen that the fluctuation of each reversing process is large; (b) is the NO_x concentration after the feedforward compensation, it can be seen that the full compensation of the disturbance can be realized under the situation that the feedforward model is perfectly matched and the control channel model is ideal linear; (c) is the change of ozone output in feedforward control.

IV. ENERGY SAVING EFFECT

Local amplification of ozone output of single reversing process in Figure 12(c) and get the following figure.

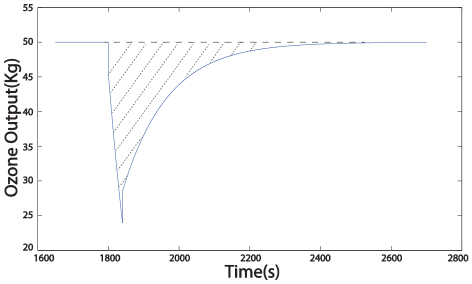


Fig.13 Ozone generator power during reversing process

The shaded area in the above figure is the saved amount of ozone Q in a single reversing process.

The ozone generator adopts CF-G-2-50 unit with air source and frequency conversion control, and its operating parameters are as follows:

TABLE II. OPERATING PARAMETERS OF OZONE GENERATOR

Parameter	Unit	Value
ozone output	$Kg\cdot h^{-1}$	50(max)
Volume	$Nm3\cdot h^{-1}$	1292-1938
Ozone concentration	wt%	3%
Power(P)	$Kwh\cdot kgO_3^{-1}$	17

The cost savings in a single day:

$$W=Q*P*W_e*K \tag{6}$$

Where W_e is industrial electricity, about 0.8 RMB /Kwh; K is the number of times of reversing in a day, calculated as reversing once half an hour, that is K=48; then calculate the area of the shaded part of Figure 9 by numerical integration method, we obtain=0.9301.

We got W=607.17 at last, the annual cost savings up to several hundred thousand RMB, it's of great significance for the enterprise.

V. CONCLUSION

Aiming at the reversing process of coking, the effect of NO_X concentration was analyzed combined with the formation mechanism of NO_x, and the correctness of theoretical analysis was verified by the data diagram; proposed a method using the response curve of continuous pulse signal connected with first order inertia transfer function to model and identify the inlet and outlet NO_x disturbance respectively, at the same time, the relationship between the loss of NO_x concentration and the average NO_x concentration was established, as well as the relationship between the loss of NO_x concentration and the average NO_x concentration, the total amount of ozone, based on the actual data of two 55-hole and 6-meter top charging coke ovens, the validity of the model is verified by comparing the established model with the actual disturbance; in the end, the feedforward control scheme is given and the feedforward control rate of ozone is obtained, The daily saving power consumption cost is calculated by simulation, which indicates the important economic significance of the paper.

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