

Modeling of Integrated Processes for Coking Flue Gas Desulfurization and Denitrification Based on RBFNN

Yaning Li, Xuele Wang and Jie Tan

*Institute of Automation, Chinese Academy of Sciences
Beijing, China
liyanning13@mailsucas.ac.cn*

Abstract—This paper proposes an efficient modeling method based on the history running data of a coking chemical company flue gas desulfurization and denitrification integration device: construct data set according to the technology principle and corresponding data preprocessing method; make division of working conditions and reduce the sample set by means of K-Means clustering method; realize static modeling for each of the conditions based on RBF neural network. The simulation results show the effectiveness of the method and the artificial neural network model.

Keywords—coking; desulfurization and denitrification; K-means clustering; RBFNN

I. INTRODUCTION

China is the world's largest coking production country[1]. In 2014, the total annual emissions are about 920 thousand tons of nitrogen oxides and 120 thousand tons of sulfur dioxide, accounting for the total nitrogen oxides and sulfur dioxide emissions by 4.1% and 0.6%. With the formal implementation of the new environmental law in January 1, 2015, the coking industry of sulfur dioxide and nitrogen oxides emissions targets are put forward strict and clear quantitative requirements. 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 release 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 adopt process flow of wet ammonia process super turbulent desulfurization and two sections of forced oxidation of urea denitrification. But desulfurization and denitrification is a process which takes chemical reaction as core, a variety of reactions coexist and with large time delay, large inertia, strong disturbance and complex nonlinear characteristics. Conventional PID and other control algorithms are difficult to achieve the desired control effect, and the overshoot is serious, even make the system unstable. Therefore, it is necessary to adopt advanced control methods such as neural network, predictive control, robust control and so on, while the

optimization control must be built on the basis of accurate model[2], so the accurate analysis and modeling of the process is the premise to realize the precise control of the gas emission index.

Aiming at the process data of the operation of desulfurization and denitrification integrative device, constructing data set with data preprocessing steps of wavelet de-noising[3], method for determining the lag time under process characteristics, etc; In order to realize the off-line modeling of the process, the data is normalized, K-means clustering algorithm is used to divide the data set into different working conditions, for specific working conditions, the algorithm is used again to reduce data[4] for constructing the neural network training set; Based on the modeling sample set for each work condition, building models respectively by RBF neural network[5]. The paper takes denitrification process as an example to illustrate the modeling process, finally, we analyze the modeling error of desulfurization and denitrification process, and the results show the effectiveness of this method.

II. BUILDING DATA SET

A. Process flow

Flue gas from coking process is fed into waste heat recovery boiler through draught fan, and the temperature is reduced from 300°C to 160°C, 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 enter the concentrated section of desulfurization tower, after spraying and washing, cooling to 60°C, through the gas cap, gas enter the absorption section of desulfurization tower, 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. After that, sulfur dioxide is removed and purified. The liquid at the bottom of the absorption section backflow to the liquid storage tank at the bottom of desulfurization tower, in order to restore the

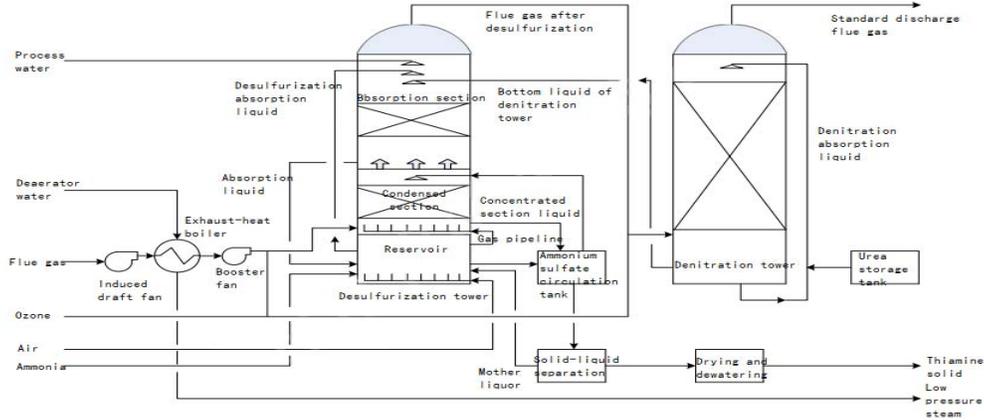


Fig.1 Desulfurization and Denitrification Integrated device for coking process

absorption capacity of the absorption liquid, ammonia have to be supplemented. Process water is sprayed from the top of desulfurization tower to keep the liquid level of the liquid storage tank 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, the ozone is mixed into the flue gas under the temperature about 60°C, part of nitric oxide in flue gas can rapidly react with ozone, converting into nitrogen dioxide. Then enter the lower part of the tower, countercurrent contacting with desulfurization absorption liquid sprayed from the top of denitrification 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 reach 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.

B. Data preprocessing

The historical data of this paper comes from the DCS system in the industrial field, and the sampling period is 1 second. Mainly includes the following variables: the level of each container variable, the key position temperature variable, pressure variable, flow variables, the PH value of absorption fluid and outlet of by-product, the concentration and flow rate of main components of the inlet of desulfurization tower and the outlet of denitrification tower.

Build data set based on the process flow as shown in table I:

TABLE I
DATA SET

Variable	Measurement units
Time	Y/M/D H:M:S
SO ₂ content of the inlet of desulfurization tower	Kg/h
SO ₂ content of the outlet of	Kg/h

denitrification tower	
NO _x content of the inlet of desulfurization tower	Kg/h
NO _x content of the outlet of denitrification tower	Kg/h
O ₃ content of desulfurization tower	m ³ /h
O ₃ content of denitrification tower	m ³ /h
Ammonia circulation quantity of desulfurization tower	m ³ /h
Urea circulation quantity of denitrification tower	m ³ /h
flow rate of the inlet of desulfurization tower	m/s
flow rate of the outlet of denitrification tower	m/s

1) Data de-noising

Because the data collected from the industrial field is affected by the production environment and equipment, instrument accuracy and measurement methods, there is a great noise interference, it is necessary to deal with the noise data before the following analysis and modeling. Wavelet transform has a good time-frequency localization characteristics, can be very good to focus on any details of the signal, such as edges, spikes, breakpoints, etc[3]. In this paper, the Sym8 wavelet is used to carry out the two layer decomposition of the data, we adopt heursure threshold value and soft threshold function.

The comparison of de-noised data and noisy data of some variables is shown in figure2.

2) Calculate content of controlled indicators

The SO₂ and NO_x in the original data are concentration values with measurement units of milligrams per second, the content value with measurement units of milligrams per second kilogram per hour is calculated by the following formula:

$$W(\text{kg/h}) = \pi * R^2 * V * 3600 * C / 10^6 \quad (1)$$

In the formula, W stands for the content value of controlled variable, R stands for the radius of inlet of desulfurization tower or the radius of outlet of denitrification tower, V stands for the flow rate of inlet or

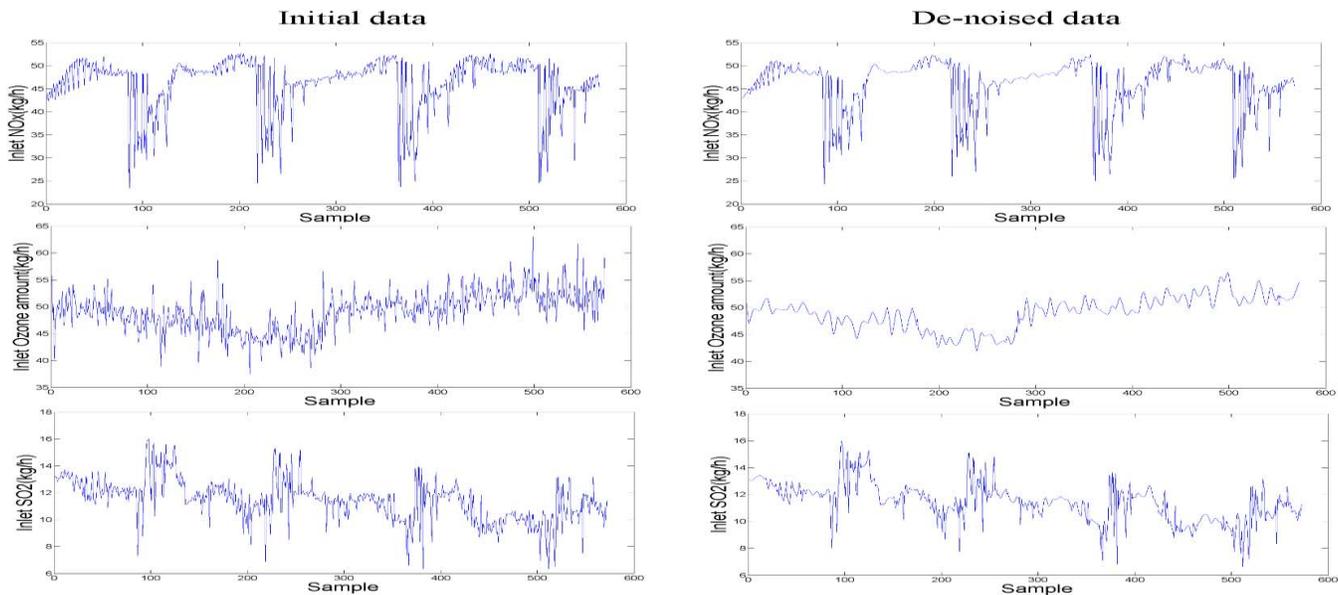


Fig.2 Comparison of wavelet de-noising

outlet of flue gas, C stands for the concentration of controlled variable.

However, the sampling period of gas flow rate is 2 seconds. So we assume that the flow rate is constant between adjacent one second, that is, we directly using industrial field data when even number of seconds, but when odd number of seconds, we use the next second data as gas flow rate.

3) Determining lag time

As the concentration detection device after desulfurization and denitrification is at the top of the denitrification tower, the concentration detection device before desulfurization and denitrification is at the central part of the desulfurization tower, and there is a large volume inside the device, the process inertia is large, so the inlet and outlet content value of flue gas in the data at the same time is not relative. The delay time of the system detection can be determined accurately by making use of the reversing process in coking production[6]: for SO_2 or NO_x , when the content of inlet and outlet reach the lowest point each half an hour, the time interval can be seemed as the delay time.

For each data set, make the relation graph between time and content for one controlled variable, using system tools to extract the time of all the inlet as well as the corresponding outlet when it reaches the lowest value. Recording as X_1, X_2, \dots, X_n and Y_1, Y_2, \dots, Y_n , so the lag time T in detection of desulfurization and denitrification process is:

$$T = \sum_{i=1}^n (Y_n - X_n) / n. \quad (2)$$

Figure out the lag time T_1 of desulfurization and the lag time T_2 of denitrification process, the mean value is the detection time lag time of this data set. Finally, move the

outlet content value of SO_2 T moment forward, as well as the content value of NO_x for each data set.

III. RBF NEURAL NETWORK MODELING

A. Modeling of denitrification process

Firstly, we take denitrification process as example to illustrate the specific process of modeling.

While in actual operation process, urea is one-time added and dissolved, and the circulation process is not interrupted, therefore, the urea is excessive, that is, excluding this variable in modeling data.

It can known from process flow that flue gas is in contact with ozone two times when it enters the desulfurization tower and the denitrification tower, but in most cases the two column ozone changes completely consistent, considering we may take strategy of controlling ozone of single tower, so in static modeling of denitrification process, the ozone variable is the sum of amount of ozone in two towers.

BP neural network model is the most commonly used models, But BP network has the limitation such as slow speed, local maximum and so on[5]. Therefore, we chose RBP network which is superior to BP network in terms of approximation ability, classification ability and learning speed.

1) Construct modeling sample set

Adjust the sampling period of 10 seconds, collecting nearly 600 sets of running data, according to analysis, the collected sample data can fully reflect the actual operation situation, so it can be used as the basic sample data. Using the observation method to eliminate the data with obvious detection error, constructing data set by the preprocessing steps described previously, then screening out variables including inlet nitrogen oxide content (kg/h), total ozone (kg/h), and export nitrogen oxides (kg/h).

In order to facilitate the learning of neural network and improve the generalization ability of neural network[7], the data in data set need to be normalized by using the min-max method[8]:

$$x^* = \frac{x - x_{\min}}{x_{\max} - x_{\min}} \quad (3)$$

In the above formula, x_{\max} and x_{\min} is the maximum and minimum values of x , x^* is the normalized data.

We choose K-means clustering algorithm[9-10] to divide the data into working conditions. The object of clustering is the 573*2 matrix of nitrogen oxides content and total ozone in the data set. Considering the number of categories K is unknown, error square sum and silhouette coefficient[11-12] is used together to determine the K. Through the enumeration method, the clustering iteration number n is 50, make the relationship diagram of error square sum and silhouette coefficient with K from 2 to 20 respectively:

From figure 3 we can see that the error sum of square decreased with the increasing of the class k, which according with the principle of the algorithm. We usually select the K value of the inflection point, that is, 3 or 4. In figure 4, the silhouette coefficient is the highest when K is 3, taking into account the actual situation of the system, we select 3 as the number of cluster categories.

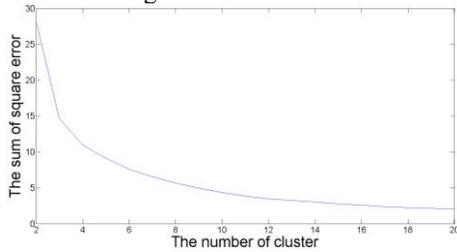


Fig.3 The relationship between the number of categories and the sum of square error

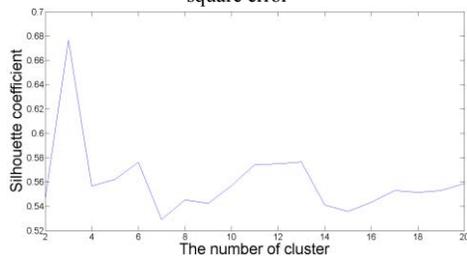


Fig.4 The relationship between the number of categories and silhouette coefficient

Clustering and drawing the cluster results as Fig.5:

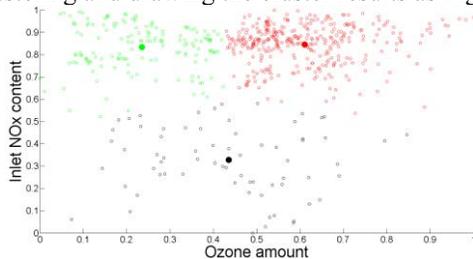


Fig.5 The clustering effect of input variables

The working conditions of the system are described in table II:

TABLE II
THE CORRESPONDING RELATION BETWEEN THE CLUSTER RESULTS AND WORKING CONDITIONS

Category color	Main operating conditions	Other description
Green	Normal state	Ozone amount [41,48]
Red		Ozone amount [48,56]
Black	Reversing process	none

To reduce the data set[4,13], each condition of the data is clustered into 30 sub categories by K-means algorithm, due to the center point of class may not be the actual data point, we select the nearest data point from the center point of each sub class as the representative of the sub class, that is, extracting the point as modeling sample. Thus, 30 data sets were extracted respectively from the original data set as the training set of three kinds of working conditions, then randomly select 10 data sets under each working condition data set as a test set of the model.

TABLE III
PART OF THE MODELING SAMPLE SET

Grade	Inlet NO _x content	Total ozone	Outlet NO _x content
1	0.6808	0.3518	0.4942
2	0.7341	0.5208	0.5668
3	0.8704	0.5764	0.7802
4	0.8544	0.4926	0.6724
5	0.8562	0.4467	0.7455
6	0.8691	0.3877	0.7266
7	0.8502	0.3391	0.7314
8	0.2992	0.4217	0.1268
9	0.3673	0.3588	0.1768
10	0.3787	0.4357	0.3182

2) RBF neural network modeling

Step 1: Take the inlet NO_x content and total ozone as input of system, outlet NO_x content as output of system, divide the sample set into input sample P and target vector T.

Step 2: Design radial basis function network, while the fitting ability and generalization ability of network is directly affected by the two parameters of the function: goal (target error) and spread (radial basis function dispersion). Here, we take a dynamic way to change these two parameters[14], take the maximum number of samples in a relative error between the output value of the original sample network and target value for each condition as the termination condition, that is, at this time the network's generalization ability and performance is the best.

Step 3: When the RBF neural network training is completed, using the network model to simulate and test the data, the code is:

$$Y = \text{sim}(\text{net}, P);$$

$$Y1 = \text{sim}(\text{net}, P1);$$

Y1 and P1 is the input samples and the network output of test data.

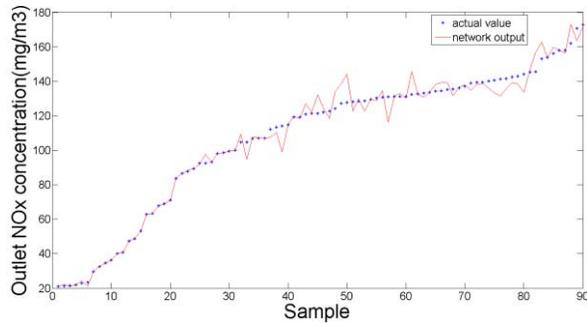


Fig.6 The comparison of output of the network and the actual value for training set

Step 4: calculate the simulation error.

The simulation error of each model after calculating as shown in table IV :

TABLE IV
MODEL SIMULATION ERROR

Model	Mean square error of training set	Mean square error of testing set	relative error of testing set
1	0.0018	0.0021	0.0603
2	0.0014	0.0021	0.0619
3	8.98e-32	0.0395	0.0683

It can be seen that the mean square error of the training set and the test set of model is ideal, and in actual production the relative error is usually less than 10%, so the network model basically meets the accuracy requirements.

B. Modeling of desulfurization process

The specific steps are the same as the modeling of denitrification process, so it's no longer repeated here .

The clustering effect of input variable is shown in figure 7:

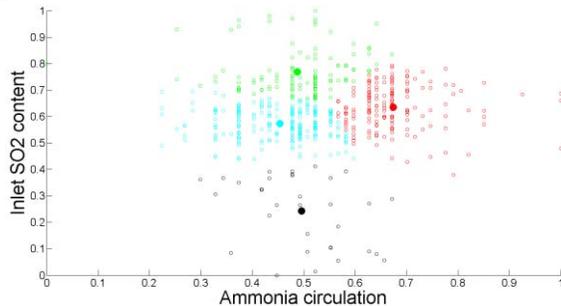


Fig.7 The clustering effect of input variables

The working conditions of the system are described in table V :

TABLE V
THE CORRESPONDING RELATION BETWEEN THE CLUSTER RESULTS AND WORKING CONDITIONS

Category color	Main operating conditions	Other description
Blue	Normal state	The circulation amount of Ammonia water is about 0.37
Red		
Green	Reversing process	
Black		

Due to the limitation of industrial field conditions, the circulation amount of ammonia water in the collected data is not changed obviously, so the system conditions are combined into two types.

Data of each condition is clustered into 30 and 60 groups by K-means algorithm respectively in consideration of the sample size, that is, extracting 30 and 60 sample sets respectively as training sets of models in two kinds working conditions, then randomly select 10 data sets under each working condition data set as a test set of the model.

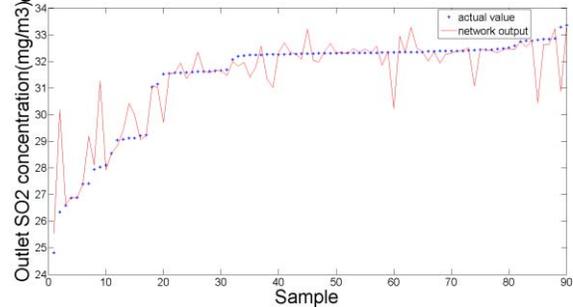


Fig.8 The comparison of training set output of the network and the actual value

The simulation error of each model after calculating as shown in table VI:

TABLE VI

MODEL SIMULATION ERROR

Model	Mean square error of training set	Mean square error of testing set	relative error of testing set
1	0.0216	0.0030	0.0638
2	0.0383	0.0191	0.0730

It can be seen that the training set's and the test set's mean square error and relative error of the model are slightly larger than the denitrification process, but the overall network model basically meets the accuracy requirements.

IV. RBF NEURAL NETWORK MODELING

The paper based on the operation data of the integrated device of flue gas desulfurization and denitrification, building data set according to the technological process and data pre-treatment method; K-means clustering algorithm is used to realize the classification of working conditions and the reduction of the dataset of modeling samples to construct the neural network training set; establishing three RBF neural network models with 2-30-1 structure of denitrification process as well as two RBF neural network models with 2-8-1 and 2-60-1 structure of desulfurization process. The simulation results show that this neural network has a good modeling effect, laying a foundation for optimal control of smoke emission index and online prediction of sulfur dioxide and nitrogen oxide

ACKNOWLEDGMENT

This work was financially supported by the National Natural Science Foundation of China (U1201251).

REFERENCES

- [1] Wang J. Strategic analysis of supply and demand of coking coal resources in China[J]. *Coal Economic Research*, 2007(10):pp.4-6.
- [2] Ma X, Chen X C, Wang S B. Nonlinear Modeling of The Boiler Combustion System in a Power Plant Based on RBF Neural Net Works. *Journal of Fuzhou University(Philosophy and Social Sciences)*, 2004,32(3):pp.295-297.

- [3] Tian S C. Study on modeling method based on noisy data driven in power plant boiler[D]. Henan: Henan University of Science and Technology, 2012.
- [4] Fang H Y. Data Processing Method of dimensionless[D].Chongqing: southwest university, 2009.
- [5] Xu Z, Yang Y L, Wang W H. An application of RBF neural network based on Matlab[J]. *Metallurgical Industry Automation*, 2009(6):pp.39-40.
- [6] Yu Z D, Zheng W H. *Technical Manual of modern coking production*[M]. Beijing: Metallurgical Industry Press, 2010.
- [7] Ding M T. Mathematics Models of Main Steam Pressure System and Its Intelligent Control Arithmetic. Hebei: North China Electric Power University,2001.
- [8] Chen S X. The Modeling and Control of The Penicillin Fermentation Process Based on The Radial Basis[D]. Beijing: Beijing Industrial University,2004.
- [9] LI X J, YUAN Z G. *Soft Sensor of the Material Flow from the New Suspension Preheater Kiln*[C].2011 3rd International Conference on Computer and Network Technology (ICCNT 2011), 2011:pp.557-561.
- [10] Han C J. Analysis and Control of Precalciner Kiln Parameters[J]. *Yunnan Chemical Technology*, 2010,37(35) : pp.24-28.
- [11] Wang K J, Li J, Zhang J Y, Guo L x. Experimental Comparison of Clusters Number Estimation for Cluster Analysis[J]. *Computer Engineering*, 2008,34(9):pp.198-202
- [12] Wang K J, Li X. Clustering algorithm selection based on the index of validity[J]. *Journal of Sichuan Normal University(Natural Science)*. 2011, 34(6):pp.915-918.
- [13] HU X H, DB-HReduction: A Data Preprocessing Algorithm for Data Mining Applications[J], *Applied Mathematics Letters*,vol.16,PP.889 — 895,2003.
- [14] Li G X, Zhang S Y, Simon O K, Wang R L. Radial Basis Function Network for Analysis of Chrome Data[J]. *Journal of Shandong Metallurgy*, 2004,26(2):pp.45-47.