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Application of NO Reduction Dynamical Model in Selective Non-catalytic Reduction Denitration System Based on Biomass

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Abstract: *In recent years, studies on denitration by applying biomass reburning method has drawn the attention of many researchers due to the characteristics of low sulfur and nitrogen content, high volatile, high ash focal activity, zero* CO_2 *net emissions, etc. Based on Chemkin software and selective non-catalytic reduction (SNCR) denitration chemical kinetic model, this paper conducted SNCR denitration chemical kinetic modeling. And the results showed that: with the increase of residence time, under the different initial concentration of NO, SNCR denitration efficiency tends to stabilize after the first increase. Moreover, the higher the initial concentration of NO, the longer the residence time which is required to achieve the greatest denitration efficiency. With the increase of ammonia nitrogen ratio, SNCR denitration efficiency increases step by step. When the normalized stoichiometric ratio (NSR) is greater than 1.5, the denitration efficiency is at a basic stable state. Under the same conditions, simulation results of the SNCR results agree well with the test results. Therefore, it can be concluded that carrying out the SNCR denitration chemical dynamics simulation using Chemkin software can provide a reference for tests and mechanism researches on SNCR, biomass reburning and advanced reburning denitration.*

Keywords: *Biomass, NO, Selective non-catalytic reduction, Denitration, Additional agent, Reduction kinetics.*

Introduction

Biomass energy is a renewable energy source derived from solar energy [2, 13]. There are a great variety of biological resources in the world including core wood, agricultural and forestry crops, bagasse, aquatic plant, municipal solid waste, domestic sludge, livestock manure etc [9]. Biomass energy accounts for 14% of total world energy consumption, ranking the fourth (after oil, coal and natural gas). China is an agricultural country with rich biomass resources. There is an annual production of more than 600 million tons of crops straws and about 400 million tons of them can be used as energy [14]. China's total forest biomass is about 19 billion tons, and the available amount is 900 million tons, and a total quantity of about 300 million tons can be used as energy. Therefore, the development potential is huge [7]. Since biomass has the characteristics of low sulfur, low nitrogen and high volatile, high ash focal activity and net zero CO_2 emissions; by using biomass to replace traditional energy sources such as coal, oil, not only can the SO_2, NO_x and CO_2 emissions of pollutants be reduced, but the sodium, potassium and other components in the ash content have auxo-action on NO_x. Therefore, using biomass as a kind of reburning fuel is a good choice [3].

In the process of boiler combustion, there are many factors which influence the formation and reduction of NO_x. Studies on this aspect are mainly experiments and numerical simulation. Because of various objective constraints, the experimental study is not only laborious but some conditions and parameters are difficult to obtain. With the birth and development of high speed large capacity computers, using computer to carry out the numerical simulation of complex combustion process has become possible. Currently, in view of the fluid flow problems, computational fluid dynamics softwares which are widely used include Huent and Phoenix, while Chemkin chemical kinetics software is frequently used for chemical reaction problems with combustion [6, 16]. Subject to the limitation of actual calculation conditions and the models, simplified process must be carried out on flow and chemical reaction during the research. One is the simplification of mixed flow process; another is the simplification of chemical kinetics reaction steps. In this chapter, on the basis of experimental study, SNCR process is taken as an object, and NO reduction dynamics simulation is carried out under typical operating condition. Then, a comparing analysis of simulation results and experimental results is conducted, which aims to provide a reference for optimization of SNCR process.

Introduction of chemical kinetic modelling

Basic approach of chemical kinetic modelling

Fuel combustion is an extremely complex process of physical and chemical changes, and there are many technical difficulties at present in applying the mathematical simulation method to simulate the whole process. Considering that the chemical reaction plays a key role in combustion process, applying method of chemical dynamics is a research direction on simulation of the combustion process. NO_x is one of the main products of the combustion process, and its generation is along with the combustion process. Therefore, simulation of NO_x formation process is inseparable from that of the whole combustion process. Here are three methods of simulation of NO_x formation.

Method one: simulate the combustion process from the aspects of air dynamic field. Due to the complex heat and mass transfer processes and aerodynamic hybrid processes of each fraction (oxidant, fuel and combustion products) during the combustion process, careful consideration can not be realized on formation of NO_x and fuel combustion chemical kinetics mechanism. Therefore, simulation on the combustion and NO_x formation process can only be regarded as a whole, that is to say, considered by the contractor reaction. Obviously this simulation cannot reflect the change process of intermediate products, nor can it analyze the main factors of NO_x formation and decomposition.

Method two: simulate the combustion process from the aspects of chemical reaction kinetics mechanism, which is different from method one since it carries out large simplification on flow process and mixing process. For example, suppose that chemical reactions occur at specific reactors – plug flow reactor (PFR) or perfectly-stirred reactor (PSR). It covers all chemical primitive reactions of all components and it can determine concentration of intermediate product and end-product at any time during the process of simulation. Through detailed analysis of the reaction process, key reaction mechanism and main influence factors which influence the formulation of NO_x can be determined. The model in this method only considers chemical primitive reaction, without involvement of influence of mixing and flow process on combustion and NO_x formation.

Method three: considering the computer hardware and software’s shortcomings, large simplification is carried out on the flow and mixing process of method one and chemical reaction mechanisms of method two, and the results obtained were integrated. In short, method three is a compromise of the former two methods. Combining with the SNCR denitration process characteristics and practical considerations of chemical dynamics research, this chapter applies the second method and builds the chemical kinetic model of NO reduction under SNCR condition and carries out relevant simulation work.

Introduction of Chemkin software

Chemkin software is a very powerful software package which can solve complex chemical reaction problems and is often used for the simulation of combustion process, catalytic process, chemical vapor deposition (CVD), plasma and other chemical reactions [15]. American National Laboratory developed and launched first generation Chemkin (Chemical Kinetics) series software in 1980, namely, the large gas-phase reaction kinetics software package, which aims to calculate and handle relevant chemical problems involved in the combustion [10]. Because the package has advantages of reasonable structure, easy to transplant, and good reliability, it is regarded as the preferred computational tool in the field of combustion simulation. Composition and structure of the software are as follows:

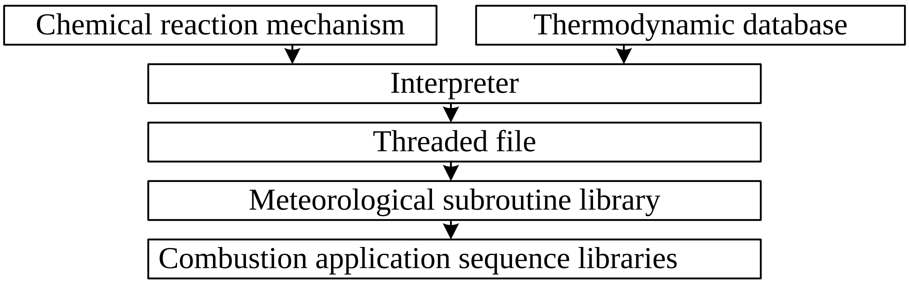

Fig. 1 Structure of Chemkin system

As can be seen from Fig. 1, the success of the software operation must be based on chemical reaction mechanism and the thermodynamic database. The interpreter is the bond of the two, which applies thermodynamic information of the corresponding component extracted from the thermodynamic database to chemical reaction mechanism involved in the combustion process. The chemical reaction mechanism is mainly composed of the following sections: elementary reaction, kinetic parameters and third body of reaction. Thermodynamic database is drawn up according to certain format, which almost covers thermodynamic information of all intermediates involved in the combustion reaction process. The threaded file is a file package which contains the coherent elements, reacting matter and the related reaction involved in the mechanism and it is the product of connector operation process. The gas phase subroutine library is a collection of a series of highly modular subroutines, which not only gives the parameters of elements, physical and chemical reactions, but also provides the state equation, thermodynamic properties of reactants, chemical reaction rate of each equation of related reaction and the corresponding material sensitivity analysis results. The corresponding output results can be applied to the flow combustion simulation process which the users are interested in.

Based on three core packages of Gas-phase Kinetics, surface dynamics and transfer process, Chemkin software provides a series of common chemical reaction models and the corresponding post-processing procedures. Gas-phase Kinetics includes composition of gas phase composition, gas phase chemical reactions and related Arrhenius data information and it is the foundation of all program calculation [12]. Surface Kinetics provides all information needed for reaction of two parties, such as surface structure of reactants, surface and internal components of reactants, etc [8]. Transport provides viscosity, thermal diffusion coefficient and the coefficient of heat conduction of the reactant components [11].

Since Gas-phase Kinetics defines thermodynamics related information of each reaction components, the operation of surface dynamics and transport must be based on it. Gas-phase Kinetics, Surface Kinetics and Transport provide the basic information of the related chemical reaction, and the dynamic link library generated can be used for subsequent procedure calls. The users can not only use build-in response model of Chemkin to achieve corresponding purposes, also they can write their own program to achieve specific functions.

Kinetic model of NO reduction

Chemical reaction kinetics model

The chain combustion mechanism is a collection of a series of reversible elementary reactions, which can be described through dynamics system of combustion process, given as follows [1]:

$$\sum_{n=1}^{N} y_{mn} Y_n = 0, \ m = 1, 2, \ldots, M. \tag{1}$$

In the above equation, Y_n is the concentration of the n^{th} component; M is the total number of primitive reaction equation in dynamics mechanism; N is the total number of components in the reaction system; y_{mn} is the stoichiometric coefficient; in component Y_n, y_{mn} participates in the m^{th} reaction. $y_{mn} > 0$ means that Y_n is formed in the m^{th} reaction. On the contrary, when $y_{mn} < 0$, Y_n will be consumed.

Stoichiometric coefficient y_{mn} is determined according to the condition of conservation of atomic number of each element which participates in the m^{th} reaction, and forms a coefficient matrix as follows:

$$\sum_{n=1}^{N} y_{mn} R_{nj} = 0, \ j = 1, 2, \ldots, J, \quad m = 1, 2, \ldots, M. \tag{2}$$

In the above equation, R_{nj} is the atom number of element j of the Y_n component; J is the number of chemical elements considered in the combustion ratio. Then reaction speed V_m of the m^{th} elementary reaction can be described as follows:

$$V_m = K_{+\mathrm{m}} \prod_{n=1}^{N} [Y_n]^{|-y_{nm}|} - K_{-m} \prod_{n=1}^{N} [Y_n]^{|+y_{nm}|}. \tag{3}$$

In the above equation, K_{+m} and K_{-m} are respectively positive and reverse rate constant of the m^{th} reaction; and reaction rate constant is determined as follows:

$$K = RT^{\beta} \exp(-E / PT). \tag{4}$$

In the above equation, R is the pre-exponential factor, β is the temperature exponent, E is the activation energy and P is universal gas constant.

Since reactant concentration is a function of temperature and pressure, considering the state function, concentration $[Y_n]$ of the n^{th} component at i moment can be determined as follows:

$$[Y_n]_i = [Y_n]_0 \frac{T_0}{T_i} \frac{\rho_i}{\rho_0}. \tag{5}$$

In the above equation, T_0, ρ_0 and $[Y_n]_0$ are respectively the temperature, pressure and concentration of the n^{th} component under standard condition.

In the general case, the overall chemical reaction rate depends not only on the speed of a single chemical reaction, but also on the reaction rate $V^{(n)}$ of a component. Suppose each primitive reaction is carried out independently, thus reaction speed of component $[Y_n]$ equals to the algebra of composition formation and consumption rate in all reactions, which is:

$$V^{(n)} = \sum y_{nm} V_m \text{ , } n = 1, 2, \ldots, N. \tag{6}$$

In the above equation, V_m is determined by Eq. (3). According to Eq. (2), J element equilibrium equations are contained in the above N differential equations as follows:

$$. \sum_{n=1}^{N} R_{nj} V^{(n)} = \sum_{n=1}^{N} R_{nj} \sum_{m=1}^{M} y_{nm} V_m = \sum_{m=1}^{M} \left(\sum_{n=1}^{N} R_{nj} y_{nm} \right) V_m = 0 \tag{7}$$

Since the reaction rate is a function of temperature and pressure, to make the equation closed, differential equation of $dT / d\iota$ and $d\rho / d\iota$ must be obtained, which is:

$$\frac{dT}{d\iota} = f_T(\iota) , \tag{8}$$

$$\frac{d\rho}{d\iota} = f_\rho(\iota) . \tag{9}$$

Therefore, mathematical model which describes the reaction system from Eq. (1) can be boiled down to solution of $N + 2$ general rigid differential equations with initial conditions as follows:

$$V^{(n)} = \sum_{m=1}^{M} y_{nm} V_m, \ \ n = 1, 2, \ldots, N - J \ \ , \tag{10}$$

$$\sum_{n=1}^{N} R_{nj} V^{(n)} = 0, \ \ j = 1, 2, \ldots, J \ \ , \tag{11}$$

$$\frac{dT}{d\iota} = f_T(\iota) , \tag{12}$$

$$\frac{d\rho}{d\iota} = f_\rho(\iota) . \tag{13}$$

The initial conditions are as follows:

$$\left[Y_n(\iota_0) \right] = \left[Y_n \right]_0 , \tag{14}$$

$$T(\iota_0) = T_0 , \tag{15}$$

$$\rho(\iota_0) = \rho_0 . \tag{16}$$

Description of chemical reaction mechanism

Specific to the SNCR process, researchers have proposed a number of reaction mechanism models. Due to different research objectives, components, categories of elementary reaction and kinetic parameters applied by various mechanism models are different, which results in great difference of accuracy on the simulation of SNCR process. The mechanism model of SNCR simulation in this chapter is designed specifically for SNCR process development. It is composed of 57 chemical reaction equations and 23 reactive substances, at the same time, it also includes the rate factor expressions and thermodynamic parameters related to each reaction material. The mechanism is generally applicable to 1000-2500 K reaction temperature and 1-10 atm reaction pressure. In the application of the mechanism, reaction conditions should not exceed the above scope, otherwise the simulated results do not have rationality. The simulation work is carried on with Chemkin4.1 software (supported by College of Energy and Mechanical Engineering, Shanghai University of Electric Power).

Computational physics model and its assumptions

In the experiment, ammonia agent nozzle uses a specially designed device in order to ensure a good mixture of medium and the main flue gas. In order to verify the applicability of the mixing situation and the reaction model, a serial model of perfectly stirred reactor and plug flow reactor in Chemkin software package is applied to perform simulation [4, 5].

SNCR physical model: By applying the above SNCR mechanism and Chemkin software, simulation of influence of different factors on the SNCR process, simulated flue gas composition (volume fraction) of the primary zone is as follows:

$$\varphi_{\mathrm{NO},in} = 200\text{-}600\,\mu\mathrm{L/L},\ \varphi\mathrm{CO_2} = 12\%,\ \varphi\mathrm{O_2} = 4\%,\ \varphi\mathrm{H_2O} = 4\%.$$

Nitrogen gas is a balance gas, and the physical model is a series reactor of PSR-PFR. PSR reactor plays a role of simulation of flue gas premix. PFR reactor is the reaction zone of SNCR and the reducing agent (ammonia) is added directly here. To conform to the reaction requirements of the actual test device, the length of PFR model is set to be 60 cm and its diameter is ϕ = 4.5 cm; reaction temperature range is 850-1150 °C, quantity of steady flow is 20 L/min (298K, 1 atm), residence time is about 0.6 second. And the reactor is set with constant wall temperature, heat insulation and ordinary pressure.

Calculation physical model assumption: In order to achieve the purpose of simulation of SNCR denitration process, corresponding assumptions are made on denitrification reactor and its internal physiochemical process as follows:

- Assume uniform mixing of gas in the SNCR reactor, and the related thermodynamic parameters of the gas mixture are identical.
- Assume that the reaction product conversion rate is determined by chemical reaction rate, rather than diffusion rate or turbulent mixing in the mixing process.
- Assume that the speed of SNCR reactor is evenly distributed on radial direction and there is no change of component concentration; chemical reaction rate only changes on axial direction, and it has nothing to do with spatial position.
- Assume that various components of a chemical reaction comply with the ideal gas state equation.
- Assume influence of the gas temperature rise in the heating process is neglected.

SNCR simulation results and analysis

Impact of reaction temperature on SNCR denitration efficiency

Fig. 2 shows the impact of reaction temperature on SNCR denitration efficiency under the condition of experiment and simulation. As can be seen from Fig. 2, with the increase of reaction temperature, SNCR presents the trend of declining after the first rise, and its best denitration is obtained at the temperature of 95 °C, among which, best denitration efficiency under the experimental conditions was 73.5%. While the analog value is 80.6% and it is slightly higher. If SNCR window temperature is defined with denitration efficiency greater than 60%, then under the condition of simulation and experiment, basic coincidence of temperature window of SNCR denitration can be realized. The denitration temperature window of the former is about 890-1060 °C while that of the later is about 850-1030 °C. Through further analysis of Fig. 2, we know that when the reaction temperature is less than 850 °C, SNCR denitration efficiency is close to zero under simulated conditions.

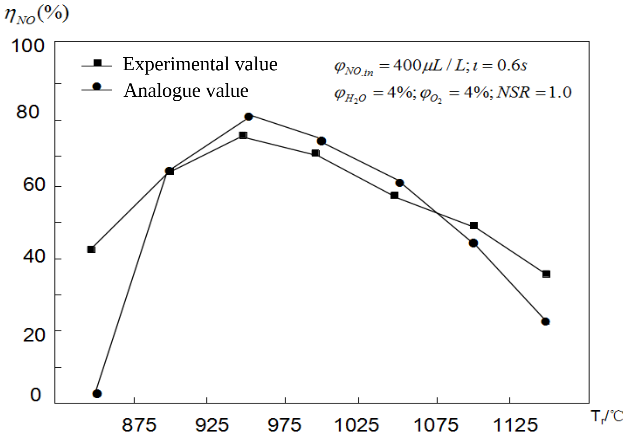

Fig. 2 Effect of reaction temperature on NO reduction efficiency

Impact of residence time and NO initial concentration on SNCR denitration efficiency

Fig. 3 shows the impact of residence time and NO initial concentration on SNCR denitration efficiency under simulated conditions. As can be seen from Fig. 3, under different initial concentration of NO, with the increase of residence time, SNCR denitration efficiency presents a stable trend after the first rise. When $\varphi_{\text{NO},in} = 200\ \mu\text{L/L}$, the time needed to achieve the maximum value of SNCR denitration efficiency is shortest with 0.24 s; when $\varphi_{\text{NO},in} = 400\ \mu\text{L/L}$, the time to achieve the maximum value of SNCR denitration efficiency is 0.4 s. The simulation time is 0.2 s less than experiment time, and it is probably because degree of mixing of simulation process is better than that of experimental process and simulation process is with the addition of water. When $\varphi_{\text{NO},in} = 600\ \mu\text{L/L}$, the time needed to achieve the maximum value of SNCR denitration efficiency is the longest with 0.92 s. In conclusion, the initial concentration of NO has a great effect on the residence time which is needed to achieve the maximum value of SNCR denitration efficiency. The higher the initial concentration of NO, the longer the residence time will be.

From further analysis, within the scope of $\varphi_{\text{NO},in} = 200\text{-}600\ \mu\text{L/L}$, with the increase of initial concentration of NO, biggest denitration efficiency of SNCR increased and the trend slowed down gradually. For example, when $\varphi_{\text{NO},in}$ increased from $200\ \mu\text{L/L}$ to $400\ \mu\text{L/L}$, the biggest denitration efficiency increased by 13.2% from 67.8% to 81.0%; while when $\varphi_{\text{NO},in}$ increased from $200\ \mu\text{L/L}$ to $600\ \mu\text{L/L}$, the biggest denitration efficiency increased by 5.4% from 81.0% to 86.4%. The denitration efficiency of simulation process is 10% higher than that of the experimental process, which may be due to the addition of water in the simulation process that promotes the denitration process. The simulation results show that denitration efficiency increased with the increase of initial concentration of NO and the increase become evident especially at low initial volume concentration of NO. However, when NO initial volume concentration reaches a certain degree, the increase trend of denitration efficiency slows down gradually.

Impact of normalized stoichiometric ratio (NSR) on SNCR denitration efficiency

Fig. 4 shows the impact of normalized stoichiometric ratio on SNCR denitration efficiency under simulation condition, from Fig. 4, it can be seen that with the increase of NSR, SNCR denitration efficiency increases gradually. When NSR increases from 0.5 to 1.0, denitration efficiency increases from 48.5% to 80.8%; when NSR increases from 1.0 to 2.0, denitration efficiency increases merely from 80.8% to 87.7%. Moreover, though continue to increase the NSR, the denitration efficiency increase is not obvious and it is basically stable at around 88.0%.

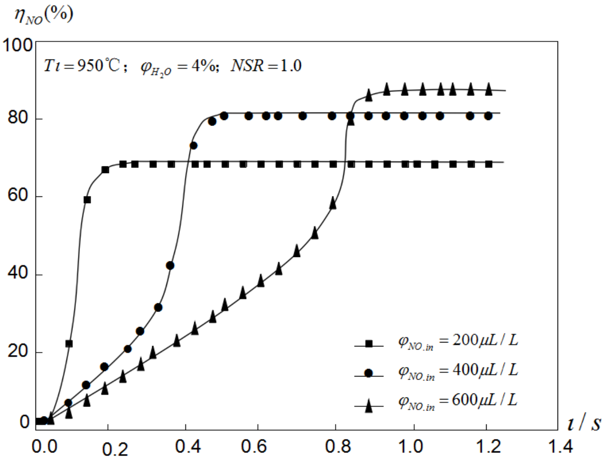

Fig. 3 Effects of residence time and initial NO concentration on NO reduction efficiency

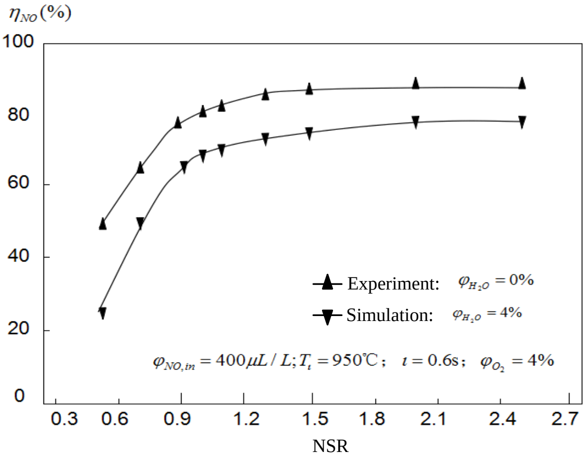

Fig. 4 Effect of NSR on NO reduction efficiency

At the same time it can be seen that the change trends of simulation results and experimental results are basically identical, since the denitration efficiency presents a gradual stable state after a rise with the increase of NSR, and the turning point is around NSR = 1.0 in both situations. However, under the same conditions, there exists a difference on denitration efficiency value between experimental results and simulation results. The biggest value of simulation of denitration efficiency is 10% higher than that of the experimental value. This can be due to the lack of water vapor in the experimental process and the fact that the degree of mixing in the simulation process is better than in the experimental process.

Conclusion

Based on the SNCR physical model assumptions, this paper carried out the simulation study on SNCR denitration, and analyzed the simulation results with the test results and obtained the following conclusions:

- Under the experiment and simulation situation, with the increase of reaction temperature, SNCR presents the trend of declining after rise in both situation and best denitration temperature is obtained at 95 °C and the denitration temperature window basically tallies. When the reaction temperature is less than 85 °C, under the condition of simulation SNCR denitration efficiency is close to zero.
- Under the different initial concentrations of NO, with the increase of residence time, SNCR denitration efficiency presents a stable trend after the first increase. The initial concentration of NO has great impact on residence time which is required for achieving the biggest denitration efficiency. The higher the initial concentration of NO, the longer the residence time will be.
- With the increase of NCR, SNCR denitration efficiency increased step by step, when NSR is greater than 1.5, denitration efficiency approximately trends to be stable.

The simulation results presented the same rule with experimental results, but the denitration efficiency of simulation is 10% higher than that of the experiment.

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