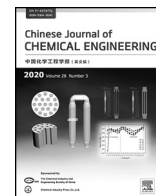




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## Article

## Dynamic optimization oriented modeling and nonlinear model predictive control of the wet limestone FGD system

Lukuan Yang, Wenqi Zhong<sup>\*</sup>, Li Sun, Xi Chen, Yingjuan Shao

Key Laboratory of Energy Conversion and Process Measurement and Control Ministry of Education, School of Energy and Environment, Southeast University, Nanjing 210096, China

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## ABSTRACT

Nonlinear model predictive control (NMPC) scheme is an effective method of multi-objective optimization control in complex industrial systems. In this paper, a NMPC scheme for the wet limestone flue gas desulphurization (WFGD) system is proposed which provides a more flexible framework of optimal control and decision-making compared with PID scheme. At first, a mathematical model of the FGD process is deduced which is suitable for NMPC structure. To equipose the model's accuracy and conciseness, the wet limestone FGD system is separated into several modules. Based on the conservation laws, a model with reasonable simplification is developed to describe dynamics of different modules for the purpose of controller design. Then, by addressing economic objectives directly into the NMPC scheme, the NMPC controller can minimize economic cost and track the set-point simultaneously. The accuracy of model is validated by the field data of a 1000 MW thermal power plant in Henan Province, China. The simulation results show that the NMPC strategy improves the economic performance and ensures the emission requirement at the same time. In the meantime, the control scheme satisfies the multi-objective control requirements under complex operation conditions (e.g., boiler load fluctuation and set point variation). The mathematical model and NMPC structure provides the basic work for the future development of advanced optimized control algorithms in the wet limestone FGD systems.

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## 1. Introduction

The wet limestone flue gas desulphurisation (WFGD) system plays an important role in air pollution prevention by offering an essential means to effectively remove sulfur dioxide ( $\text{SO}_2$ ) from flue gas in coal-fired power plants [1]. However, the WFGD system is a complex, large-scale industrial system, which consumes massive electric energy and plenty of desulfurizing agents. Therefore, the economic performance of the wet limestone FGD system has recently aroused increasing concern.

Generally, the dynamic optimization requirements of the wet limestone FGD control system are:

- I. Regardless of the fluctuation of  $\text{SO}_2$  concentration in original flue gas, the outgoing  $\text{SO}_2$  of the system should maintain in a reasonable range. The tracking zone of the  $\text{SO}_2$  concentration under full load is no more than  $35 \text{ mg} \cdot \text{m}^{-3}$  based on Chinese domestic legislation.
- II. For the purpose of energy saving, energy consumption becomes an irreplaceable part of control objectives.

<sup>\*</sup> Corresponding author.

E-mail addresses: [220170471@seu.edu.cn](mailto:220170471@seu.edu.cn) (L. Yang), [wqzhong@seu.edu.cn](mailto:wqzhong@seu.edu.cn) (W. Zhong), [sunli12@seu.edu.cn](mailto:sunli12@seu.edu.cn) (L. Sun), [chenxiseu@seu.edu.cn](mailto:chenxiseu@seu.edu.cn) (X. Chen), [yjshao@seu.edu.cn](mailto:yjshao@seu.edu.cn) (Y. Shao).

In order to realize these two dynamic optimization objectives, massive attempts have been made to build a proper dynamic nonlinear model that accurately reflects dynamics properties of the wet limestone FGD system. Kikkawa *et al.* [6] proposed a model which separates the wet limestone FGD system into two regions: the gas-to-liquid contact zone and the slurry tank. Based on this model, numerous modifications and improvements were developed. In the gas-to-liquid contact zone, different models were proposed based on the gas-liquid mass transfer theory, such as the model of spray scrubbing tower [2–5], the model of the wet limestone FGD system concerning the particle size distribution (PSD) of limestone [6], the model to forecast the tendency of  $\text{SO}_2$  removal efficiency [7] and the model of full-scale wet limestone FGD processes [8–9].

Based on the models of the wet limestone FGD system, numerous advanced control algorithms were applied to improve optimal control performance, such as neighboring extremal control [10], real-time optimization method [11], real-time iterations strategy [12], model predictive control (MPC) with incomplete optimization [13], real-time nonlinear optimization strategy [14], best optimal performance control [15], advanced-step control [16], fast economic model predictive control [17–18].

There is no doubt that the advanced control strategies above improve the control performance. In recent years, scholars have proposed

some nonlinear model predictive control algorithms for specific application objects [19–20]. For example, Zhang [21] developed an adaptive state feedback predictive control (SFPC) scheme and an expert control scheme to control and optimize a delayed coking furnace, which improved the performance of temperature control in normal operation. Flemming [22] investigated a multistage NMPC scheme with adaptive on-line scenario-tree generation, which increased the economic benefits without impairing the attained level of robustness for the process. Yu [23] compared the differences between economic NMPC and tracking MPC based on a solid sorbent-based CO<sub>2</sub> capture process, and proved that NMPC has a good performance on setpoint tracking and stability. Telen [24] proposed a NMPC scheme for a droop model, which reduced the economic loss by adjusting the parameters of the objective function.

Furthermore, the nonlinear model predictive control (NMPC) strategy has proved that it achieves a good economic performance on the application of complex industrial systems [25–28], e.g., refinery process, sewage treatment plant and double continuous stirred tank reactor (CSTR) reactor, but there are very few academic results reported about the NMPC application in the wet limestone FGD system. As a result, the NMPC has great potential applications on the wet limestone FGD system to improve economic performance under different working conditions.

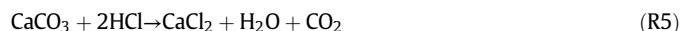
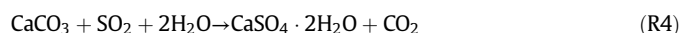
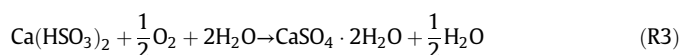
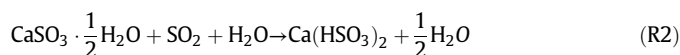
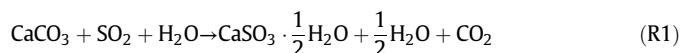
In order to realize the multi-objective optimized control of desulphurisation process, this paper presents a NMPC scheme based on the mathematical model of the wet limestone FGD system so that, i) the dynamics properties of the wet limestone FGD system can be revealed based on the model; ii) the dynamic optimization requirements consisting of set point tracking and energy saving in the wet limestone FGD system can be satisfied; iii) several advanced control and optimization methods in the paper can be verified by field data; iv) the advantages and benefits of NMPC for the wet limestone FGD system can be studied deeply in an academic point of view.

## 2. Mathematical Modeling of Wet Limestone FGD System

### 2.1. Plant description

When modeling a system for a specific purpose, the selection of variables and assumptions are critical [29]. This section attempts to put

forward a model that reflects all important dynamics properties based on the first law of thermodynamics and the law of conservation of mass. The wet limestone FGD system of a 1000 MW coal-fired power plant in Henan Province, China, is selected for the modeling validation (Fig. 1.). With the idea of block modeling, the model of the whole wet limestone FGD system is divided into several modules based on the disparities of regional peculiarity and dynamics properties in different areas [30–32]. Based on the conservation laws and the chemical equilibrium method, some reasonable parameters are selected which can reflect the chemical reaction process of the system. The chemical reaction process of removing SO<sub>2</sub> in the wet limestone FGD system contains three critical steps [33,34]. Firstly, the chemical reaction of SO<sub>2</sub> and CaCO<sub>3</sub> generates CaSO<sub>3</sub> (R1). Secondly, because of the intermediate reaction of O<sub>2</sub> and HSO<sub>3</sub><sup>−</sup> in the flue gas, partial Ca(HSO<sub>3</sub>)<sub>2</sub> is converted into CaSO<sub>4</sub>·2H<sub>2</sub>O, namely gypsum (R2–R3). The remaining Ca(HSO<sub>3</sub>)<sub>2</sub> in the slurry tank is oxidized into CaSO<sub>4</sub> by the air blasted from the oxidation fan. Of course, since the original flue gas contains other components, there are some other reactions in the chemical reaction process, such as: SO<sub>3</sub>, HCl and HF react with CaCO<sub>3</sub> and generate CaSO<sub>4</sub>·2H<sub>2</sub>O, CaCl<sub>2</sub> and CaF<sub>2</sub> (Eqs. (R4)–(R6)).



### 2.2. Assumptions

The aforesaid reaction process contains too many influencing factors, therefore it is important to propose some hypotheses for

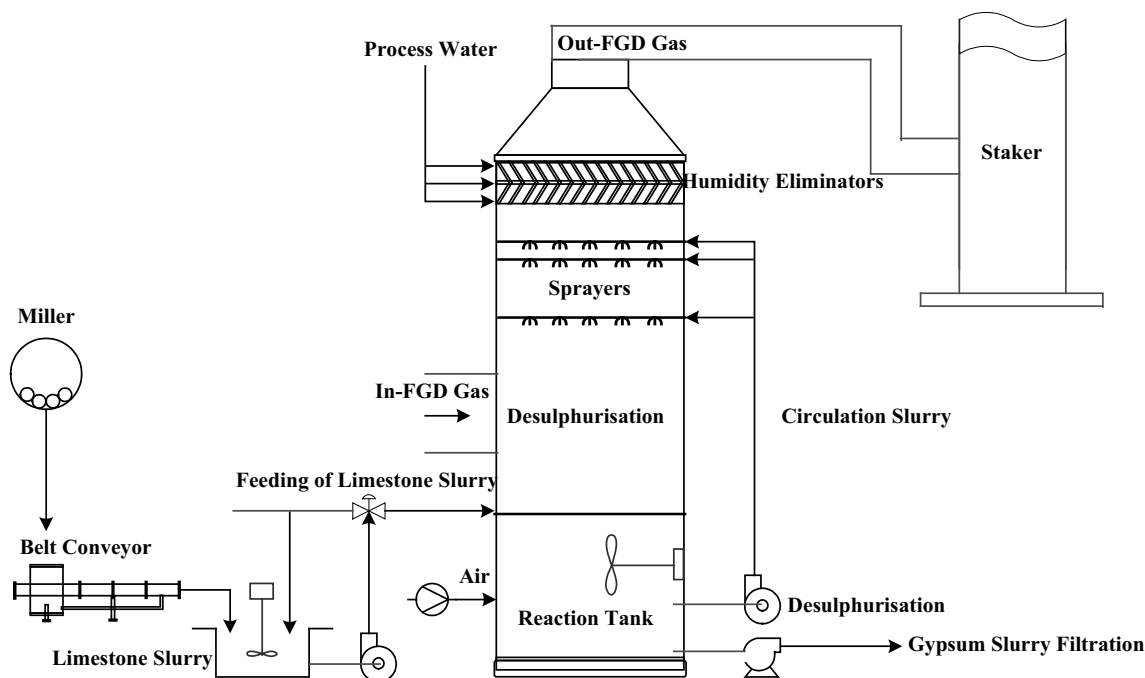


Fig. 1. A schematic view of a wet limestone FGD system.

simplification. First of all, the entire desulphurisation system is assumed to be divided into two primary modules: The first module is the gas-to-liquid contact zone which has a great impact on SO<sub>2</sub> removal efficiency and the second module is the slurry tank which has a significant influence on the pH value in this container. Secondly, the chemical reaction and mass transfer should be taken into consideration when calculating the concentration changing inside the falling droplets where the chemical reactions between different ions will be calculated comprehensively [35–37].

What's more, the two modules are connected by the circular slurry, namely, the concentration of each solute at the outlet of the slurry tank is regarded as same as the concentration at the inlet of the sprayers. Based on the field data of the wet limestone FGD system in Sanmenxia power plant, China, the accuracy of model dynamics properties is validated so that simulation experiments and analysis of dynamics properties are carried out (Table 1).

As a result, the following assumptions are proposed in this paper for the purpose of simplification:

- The time of achieving charge conservation inside the droplets is regarded as infinity so that the time of it is ignored, while the time of SO<sub>2</sub> absorption in the gas-to-liquid contact zone, SO<sub>2</sub> mass transfer in gas-liquid interface, limestone dissolution and gypsum crystallization is taken into consideration.
- Based on the primary characteristics of the two modules, the gas-to-liquid contact zone is assumed to be a plug flow reactor, and the slurry tank is assumed to be a perfectly mixing flow reactor, namely the concentration of solutes is uniform everywhere in the tank. The substance exchange between two modules is fulfilled by the circular slurry.
- The concentration of different solutes in the slurry tank is equal to the outgoing slurry concentration of the slurry tank and the concentration of circular slurry.
- Evaporation and heat transfer between flue gas and droplets are neglected.
- The shape of droplets is spherical. The concentration changing inside the droplets  $\int_0^t C_i$  is assumed to be  $\Delta C_i$  because the calculated error can be weakened by the feedback from predictive control.

### 2.3. Rate of critical reactions

Based on the foregoing hypotheses, it is known that the dynamics properties of the wet limestone FGD system is largely determined by the following four reaction rates: the rate of SO<sub>2</sub> absorption in the gas-to-liquid contact zone, SO<sub>2</sub> mass transfer in gas-liquid interface, limestone dissolution and gypsum crystallization. The interaction of these procedures is complicated so that this subsection derives a mathematical model containing the procedures above which describes dynamics properties of the system.

**Table 1**  
Description of the wet limestone FGD unit

| Parameter                                  | Value                             |
|--------------------------------------------|-----------------------------------|
| Boiler type                                | Ultra-supercritical boiler        |
| Air pollution control devices              | Fabric filter + wet limestone FGD |
| Wet limestone FGD process                  | CaO–CaCO <sub>3</sub>             |
| Inlet gas temperature/°C                   | 90                                |
| Slurry density/kg·m <sup>-3</sup>          | 1134                              |
| Gas velocity/m·s <sup>-1</sup>             | 3.5                               |
| Circular slurry residence time/min         | 5                                 |
| Gas residence time/s                       | 7.56                              |
| Flow direction (downstream/countercurrent) | Countercurrent                    |
| Desulphurisation tower diameter/m          | 23/20                             |
| Desulphurisation tower height/m            | 46                                |
| Slurry tank height/m                       | 13                                |
| Slurry tank volume/m <sup>3</sup>          | 5173                              |
| Sprayer layer number                       | 5                                 |

#### a) Absorption of SO<sub>2</sub>, CO<sub>2</sub>

According to the two-film theory, the SO<sub>2</sub> mass transfer in the gas-liquid interface of droplets can be expressed by the rate of mass transfer (Eqs. (A1)–(A5)).

#### b) Oxidation of HSO<sub>3</sub><sup>-</sup>

The oxidation fan blows air into the slurry tank to oxidize HSO<sub>3</sub><sup>-</sup> into SO<sub>3</sub><sup>2-</sup> so that the pH value is maintained within a certain range.

$$R_{ox} = K_{ox} \cdot (C_{HSO_3^-} + C_{SO_3^{2-}})^{1.5} \quad (1)$$

where  $K_{ox}$  is the constant of oxidation rate,  $R_{ox}$  is the oxidation rate,  $C_{HSO_3^-}$ ,  $C_{SO_3^{2-}}$  is the concentration of HSO<sub>3</sub><sup>-</sup>, SO<sub>3</sub><sup>2-</sup>.

#### c) Dissolve of limestone

Limestone is the desulfurizer of the wet limestone FGD system [38]. The Ca<sup>2+</sup> dissolved from limestone neutralizes the negative ions, e.g., SO<sub>4</sub><sup>2-</sup>, to adjust the pH value and accelerate the crystallization rate of gypsum.

$$R_{CaCO_3} = K_{CaCO_3} \cdot (1 - RS_{CaCO_3}) \quad (2)$$

$$RS_{CaCO_3} = \frac{C_{Ca^{2+}} \cdot C_{CO_3^{2-}}}{K_{cp}} \quad (3)$$

where  $RS_{CaCO_3}$  is the relative degree of saturation of CaCO<sub>3</sub>,  $K_{cp}$  is the equilibrium solution constant of limestone,  $C_{Ca^{2+}}$ ,  $C_{CO_3^{2-}}$  is the concentration of Ca<sup>2+</sup>, CO<sub>3</sub><sup>2-</sup>.

#### d) Crystallization of gypsum

The rate of crystallization of gypsum can be calculated by empirical formula (Eqs. (A6)–(A8)).

$$R_{gy} = K_{gy} \cdot (RS_{gy} - 1) \quad (4)$$

where  $R_{gy}$  is the crystallization rate of gypsum,  $k_{gy}$  is the crystallization rate constant of gypsum,  $RS_{gy}$  is the relative saturation of gypsum.

### 2.4. The gas-to-liquid contact zone module

In the gas-to-liquid contact zone, concentration of different solutes changes along the altitude direction (Eq. (5)). Under a steady state, formula (5) can be replaced by formula (6).

$$\frac{dC_i}{dz} = \tau_A \cdot \sum R_i \quad (5)$$

$$\Delta C_i = \tau_A \cdot \sum R_i \quad (6)$$

where  $R_i$  is the generation rate of component  $i$ ,  $C_i$  is the liquid molar concentration of component  $i$ ,  $\tau_A$  represents the time that droplet falls from the spray nozzle to the surface of the slurry tank.

Based on the assumption, the gas-to-liquid contact zone is simplified as a plug flow reactor, whose dynamics characteristics are depicted by the formulae in the Table A1. The following formulae have high accuracy on the overall behavior of the module because they can calculate the concentration of SO<sub>2</sub> and CO<sub>2</sub> in the flue gas and the concentration of different ions concentration inside the droplets precisely at any height.

However, it takes large computational cost due to partial differential equations, making it not practical for model prediction in the NMPC scheme. From the perspectives of the model prediction and optimal control, there is no need to calculate the concentration at each height, because we only focus on the concentration dynamics at the boundary. Therefore, the partial differential Eqs. (1)–(7) in Table A1 can be changed into difference equations (shown in Table A2).

Based on the formulae in Table A2, the gas-to-liquid contact zone is thus divided into several subregions to avoid solving partial equations so that computational cost is reduced (the calculation schematic diagram is shown in Fig. 2.). It is noteworthy that the ion concentration in Table A2 is related to formulae in the slurry tank module, which is changed with time and provides basic information of dynamic characteristics for the gas-to-liquid contact zone modules.

As an illustrative example, the concentration change of sulfur substance in the droplets from sprayers to the gas–liquid interface of the slurry tank is shown below. The concentration change of other substance is shown in Appendix A (Eqs. (A9)–(A14)).

$$\frac{d}{dz} (C_{SO_2} + C_{HSO_3^-} + C_{SO_3^{2-}}) = \tau_A \cdot (N_{SO_2}, A - R_{ox}) \quad (7)$$

$$\Delta (C_{SO_2} + C_{HSO_3^-} + C_{SO_3^{2-}}) = \tau_A \cdot (N_{SO_2}, A - R_{ox}) \quad (8)$$

## 2.5. Slurry tank module

The slurry tank module makes control more difficult due to its large time-delay and inertia. To overcome the control difficulty, it is significant to investigate chemical reactions of this module.

The slurry tank is regarded as a well-mixed reactor due to the influences of the agitator and forced oxidation air. Therefore, the average

concentration of the solutes in the slurry tank is assumed to be uniform and equal to the slurry concentration in the outlet of the tank and the circular slurry concentration. The following equations are deduced from the law of mass conservation and the conservation law of electric charge.

$$\frac{dC_i}{dt} = T_i + \sum R_i \quad (9)$$

$$T_i = (Q_f \cdot C_{i,f} + Q_{rf} \cdot C_{i,rf} - Q_r \cdot C_i - Q_{out} \cdot C_i) / V_T \quad (10)$$

$$C_{A^+} \cdot \frac{dC_{B^-}}{dt} + C_{B^-} \cdot \frac{dC_{A^+}}{dt} = K_i \cdot \frac{dC_{AB}}{dt} \quad (11)$$

where  $T_i$  represents the concentration changed by the flow moving into and out of the slurry tank, respectively.  $Q_f$ ,  $Q_{rf}$ ,  $Q_r$ ,  $Q_{out}$  respectively represents the flow of the fresh limestone slurry, the flow of the circular slurry, the flow coming back from the circular slurry, the flow of the draining slurry tank.  $C_{i,f}$ ,  $C_{i,rf}$ ,  $C_i$  represents the average concentration of the fresh slurry, slurry tank and the flow coming back from the circular slurry respectively.

For example, the concentration change of sulfur substance in the slurry tank is illustrated below.

$$C_{A^+} \cdot \frac{d}{dt} (C_{SO_2} + C_{HSO_3^-} + C_{SO_3^{2-}}) = T_{SO_2} + T_{HSO_3^-} + T_{SO_3^{2-}} + N_{SO_2}, T - R_{ox} \quad (12)$$

$$C_{H^+} \cdot \frac{dC_{HSO_3^-}}{dt} + \frac{dC_{H^+}}{dt} \cdot C_{HSO_3^-} = K_1 \cdot \frac{dC_{SO_2}}{dt} \quad (13)$$

By summarizing the ordinary differential equations and algebraic equations, a nonlinear mathematical model can be expressed which

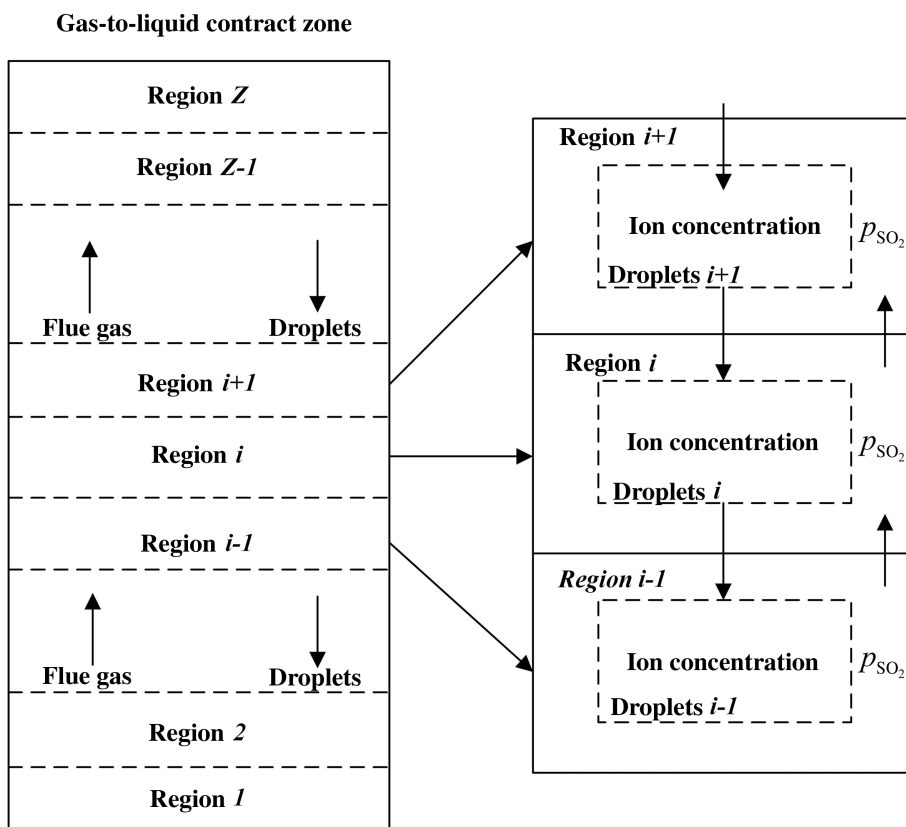


Fig. 2. The calculation schematic diagram of the gas-to-liquid contact zone.

gives profound understanding of the physical mechanisms reflecting the dynamic characteristics of the wet limestone FGD system.

## 2.6. Simulation results

Based on the Matlab/Simulink simulation environment, the lumped parameter model of the wet limestone FGD system which couples several modules together is simulated. The simulation uses a fixed time step of 1 s and is solved by the ode15s algorithm.

From the perspective of optimal control, the flow rate of the fresh limestone slurry ( $u_1$ ) and the circular slurry ( $u_2$ ) are control variables, while the  $\text{SO}_2$  removal efficiency ( $y_1$ ) and the pH value in the slurry tank ( $y_2$ ) are controlled variables. On one hand, the flow rate of the fresh limestone slurry should be adjusted duly to keep the pH of the slurry tank in a certain range, e.g., 5.2–5.7. In this subsection, a simulation experiment of the slurry volume step disturbance is carried out. Fig. 3 shows the result when the system is running with constant flow of the circular slurry (and suddenly increasing the flow of the fresh limestone slurry). At the beginning, both the efficiency of desulphurisation and the pH are in initial steady control values, then they start to increase when the flow of the fresh limestone slurry increases. It is not surprising to find that the increasing fresh limestone slurry provides more  $\text{Ca}^{2+}$ , which accelerates the crystallizing speed of gypsum. Therefore, the absorption rate of  $\text{SO}_2$  in the scrubber decreases in the influence of the decreases of  $\text{SO}_4^{2-}$ . What's more, the increase of  $\text{Ca}^{2+}$  leads to the increase of pH in the circular slurry, which will also make the absorption rate of  $\text{SO}_2$  in the scrubber higher.

On the other hand, it is necessary to adjust the circular slurry frequently based on the working conditions in order to achieve superior control behavior. When attempting to keep a constant flow of the fresh limestone slurry (and suddenly increasing the flow of the circular slurry), the simulation result is shown in Fig. 4. With the sudden increase of the flow of the circular slurry, the pH rises quickly and then slowly decreases to a new constant value. The export concentration of  $\text{SO}_2$  decreases and the efficiency of desulphurization increases to a new steady value. The increase of pH in the beginning is caused by the more absorption of the  $\text{SO}_2$ , giving a higher pH than the level before increasing the flow of the circular slurry. In the tank, the  $\text{H}^+$  then increases when  $\text{SO}_2$  is changed to  $\text{SO}_4^{2-}$ , which will slightly decrease the pH.

Under the nominal operation condition, the comparison between the simulation result of the mathematical model and the field data is shown in Fig. 5. It can be seen that the mathematical model can well simulate the dynamic characteristics of the system, indicating that the model has high dynamical accuracy.

In general, the main focus of controller design is whether the model can imitate dynamics well. The accuracy of model will be acceptable if the model correctly reflects the approximate trend of dynamics properties. The reason for the dynamic deviations in some cases may result in the fact that the temperature is not considered when calculating the absorption rate and chemical reaction rates.

## 3. Design of the Nonlinear Model Predictive Control (NMPC) structure

The aforementioned mathematical model is used to be the prediction model in MPC scheme. In MPC controller scheme, it is a modus operandi to take the value obtained from the prediction model as controlled variables [39]. Thus the MPC control structure of the wet limestone FGD system is shown in Fig. 7. In this section, the aforementioned nonlinear model is henceforth linearized into a state space model on a nominal operation condition so that the linear model predictive control (LMPC) strategy can be proposed on the basis of state space model.

### 3.1. Model linearization

The mathematical model above can capture the dynamics properties of the wet limestone FGD system well. However, it is noteworthy that the model has two severe drawbacks, making it not practical for the design of MPC control system. First, the mathematical model has various ordinary differential equations so that the solution process is time-consuming. Second, there are plenty of middle variables in the aforementioned model, which will increase the control mismatch and difficulty. In this subsection, the mathematical model is linearized based on the method of step response identification.

The nominal operating condition is chosen as the average value of controlled variables and controlled variables during three hours in a full load condition, that is,  $y_1 = 99.14\%$ ,  $y_2 = 5.54$ ,  $u_1 = 0.0194 \text{ m}^3 \cdot \text{s}^{-1}$ ,  $u_2 = 18.0556 \text{ m}^3 \cdot \text{s}^{-1}$ . Based on the step-response data, the linearized

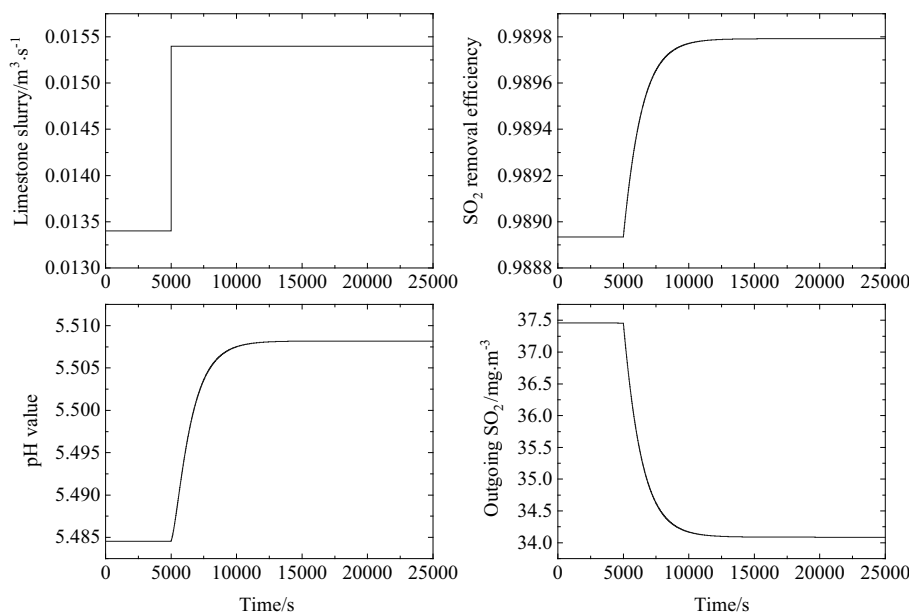


Fig. 3. Open-loop responses of the flow of the limestone slurry under a nominal operating condition ( $0.002 \text{ (m}^3 \cdot \text{s}^{-1})$  step increase in  $u_1$ ).

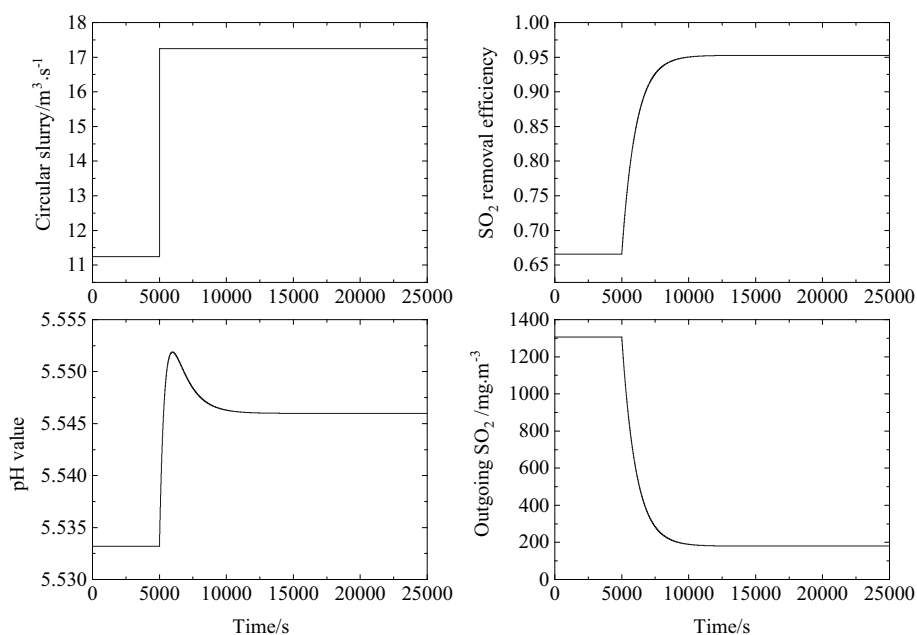


Fig. 4. Open-loop responses of the flow of the circular slurry under a nominal operating condition ( $6 \text{ m}^3 \cdot \text{s}^{-1}$  step increase in  $u_2$ ).

nominal model is consequently determined from the nonlinear model. Take  $u_1$ - $y_1$  module as an example:

$$\begin{aligned}
 g_{11}(s) &= \frac{2799s + 6.425}{10000000s^2 + 22910s + 1.327} g_{12}(s) \\
 &= \frac{25000s + 111.7}{1000000s^2 + 0.006703s + 5.631} g_{21}(s) \\
 &= \frac{46.01s + 6.065}{1000000s^2 + 2285s + 1.268} g_{22}(s) \\
 &= \frac{3861s + 1.722}{100000000s^2 + 265900s + 172.4} \quad (14)
 \end{aligned}$$

Noted that the transfer function model (Eq. (13)) is an approximation of a single operation condition. Fig. 6. shows the open-loop responses of the linearized models under three typical working conditions in Table 2.

It is vividly shown that, because of the nonlinearity of the system, the responses vary with different working conditions. To reduce prediction deviation caused by linearization, a method of multimodality will be proposed in Section 3.

### 3.2. The LMPC control structure

In this subsection, the theory and design of LMPC strategy is described. It is well known that MPC controller based on linear state space is widely applied in industry process because of its succinct definition and convenience to employ. According to the basic structure of model predictive control, the implementation of the LMPC method in the wet limestone FGD system can be divided into three main steps:

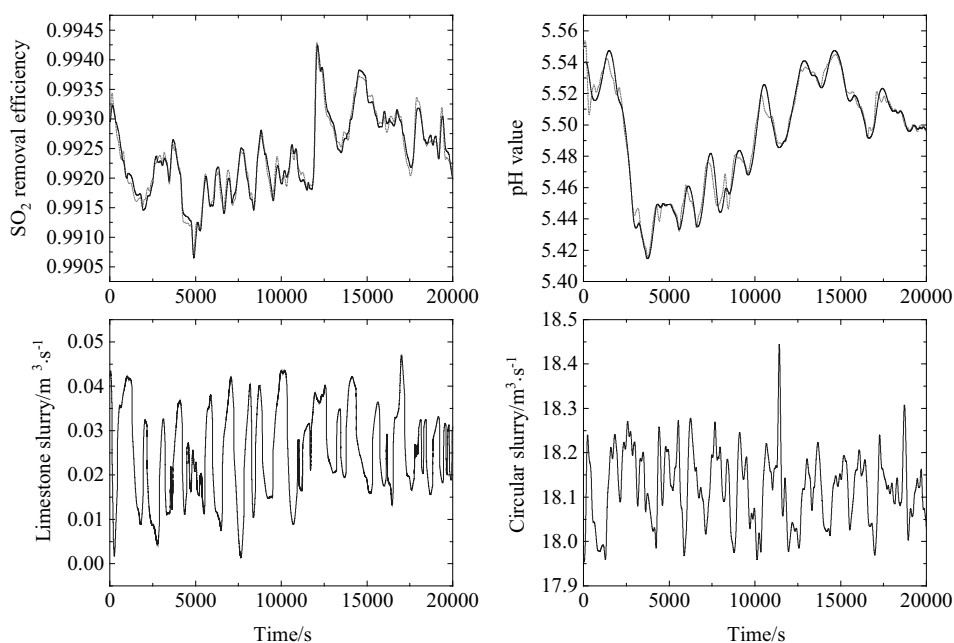


Fig. 5. Comparison results for model verification. (Red dotted line is the model output and blue solid line is the field data).

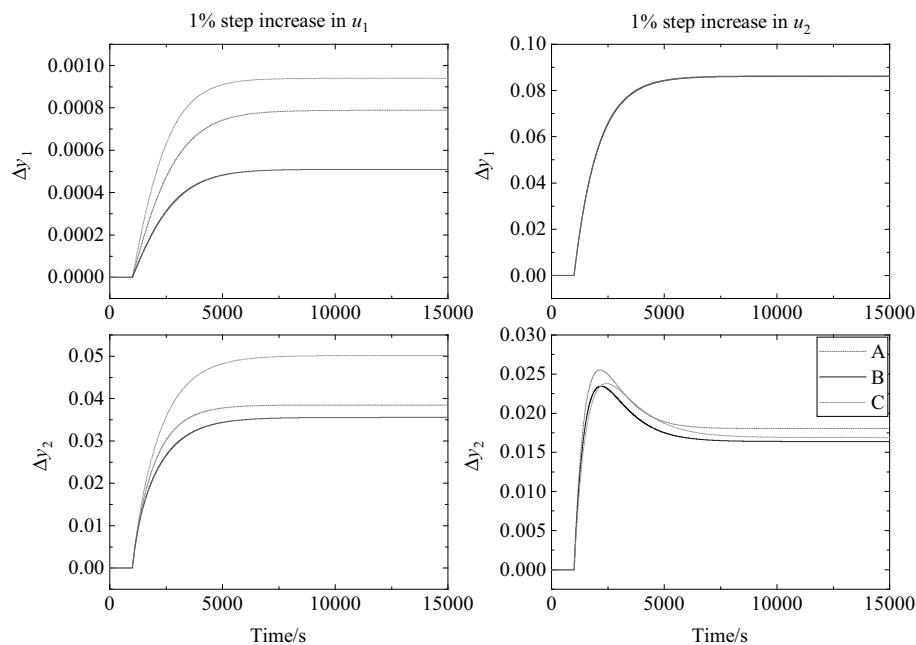


Fig. 6. Open-loop responses of three typical operating conditions.

model prediction, objective function optimization and receding horizon control.

### 3.2.1. Model prediction

In model prediction stage, it is supposed that the prediction horizon of the prediction model, namely the number of samples, is  $N_p$  in MPC. At

the sample time  $k$ , the future state variables, namely  $x(k+1|k)$ ,  $x(k+2|k)$ , ...,  $x(k+N_p|k)$ , are predicted based on the state variables measured in the current system  $x(k|k)$  and the prediction control variables in the future for  $N_p$  number of samples  $\Delta u(k+1|k)$ ,  $\Delta u(k+2|k)$ , ...,  $\Delta u(k+N_p|k)$ .  $N_p$  is the length of the optimization window, while  $N_c$  is the length of the control window [34].

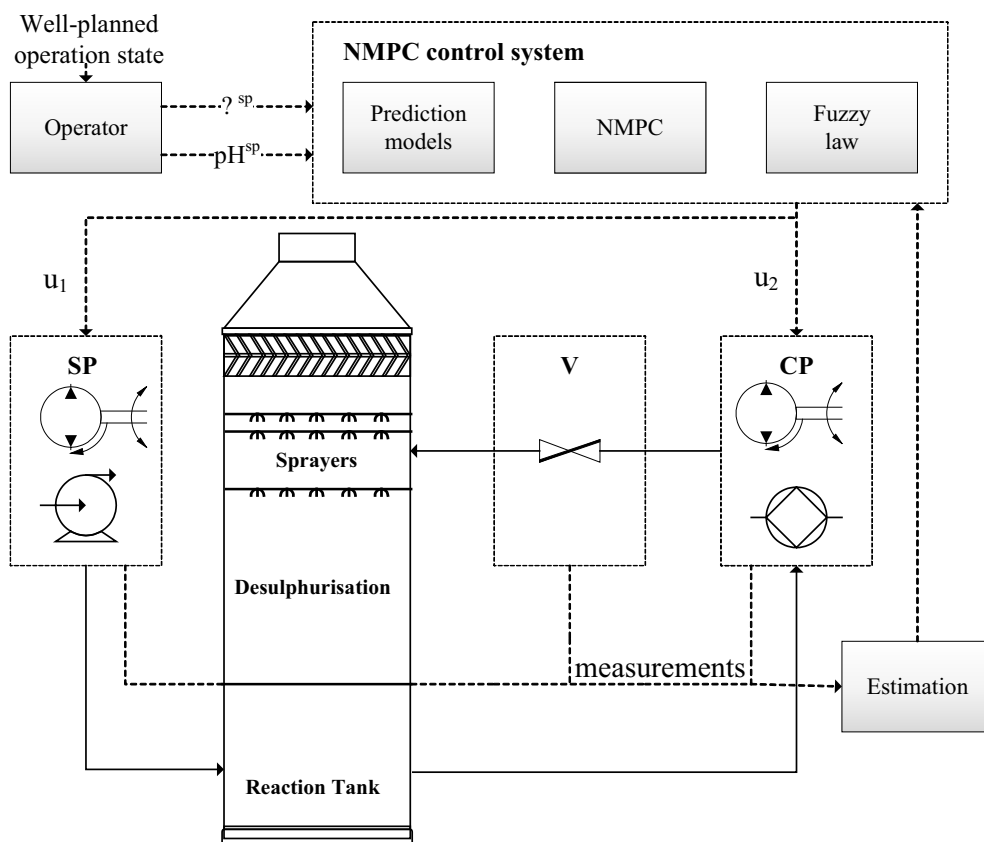


Fig. 7. Schematic of the NMPC structure used in the wet limestone FGD system.

**Table 2**

Three typical working conditions

| Working condition | $u_1/\text{m}^3 \cdot \text{s}^{-1}$ | $u_2/\text{m}^3 \cdot \text{s}^{-1}$ | $y_1$  | $y_2$ |
|-------------------|--------------------------------------|--------------------------------------|--------|-------|
| A (70%)           | 0.0154                               | 12.9833                              | 0.7831 | 5.492 |
| B (80%)           | 0.0201                               | 16.7856                              | 0.8571 | 5.567 |
| C (100%)          | 0.0227                               | 25.4883                              | 0.992  | 5.641 |

### 3.2.2. Objective function optimization

The objective of optimization is to make the controlled variables predicted by the model,  $y_1$  and  $y_2$ , as close as possible to the set-point signal given at sample time  $k_i$ . During the optimization, it is assumed that the set-point signals keep constant. Then by solving the optimization problem, the best value of control variables is obtained, which minimize the deviation between the controlled variables and the set-point signal. The optimization problem is written as minimizing the objective function in quadratic form with constraints (Eqs. (15), (16)).

$$\min_u \sum_{j=k}^{N-1} (r(j) - y_p(j|k))^2 \quad (15)$$

$$\text{s.t. } \tilde{x}(j+1) = f_d(\tilde{x}(j), u(j), 0)$$

$$\tilde{x}(k) = x(t_k)$$

$$(\tilde{x}(j), u(j)) \in Z, j = k, k+1, \dots, k+N-1 \quad (16)$$

where  $\tilde{x}(j)$  represents the predicted state variables at time  $j$ ,  $u(j)$  represents the control variables to be solved at time  $j$ ,  $r(j)$  represents the set point,  $y_p(j|k)$  represents the value of control variable,  $f_d$  represents discrete state space form of prediction model,  $x(t_k)$  represents the real value of the state variables,  $Z$  represents the amplitude constraints.

In a word, the dynamics properties from sample time  $k$  to  $k+N_p$  of the wet limestone FGD system can be calculated by the prediction model. According to the prediction model, the objective function and inequality constraints of the optimization problem can be constructed. Interior point methods, as an effective means to handle the convex optimization problem, can be used to solve the problem above.

### 3.2.3. Receding horizon control

The first vector  $u(k+1|k)$  of the predictive sequence is implemented as the control varieties of the wet limestone FGD system. When the next sample time arrives, new measured value is obtained and considered as a new initial condition to predict dynamics properties of the wet limestone FGD system in the future. The new predictive sequence is used to solve the new optimization problem so that the first sample of the new sequence can be used into the system. This procedure is repeated to achieve the receding horizon control.

### 3.3. The NMPC control structure

The LMPC strategy performs well on the linear models (Figs. 8 and 9), however it has some serious drawbacks when used in the nonlinear models. The wet limestone FGD system is highly nonlinear and has large time delay, so that the MPC controller designed on a nominal operation condition cannot achieve satisfactory control performances in different operating conditions. This subsection design a NMPC controller based on multimodality to improve the set point tracking ability of MPC (Fig. 9).

#### 3.3.1. Linearity analysis and fuzzy law

To achieve a better control performance on nonlinear systems, there have been various methods raised based on MPC [40]. In the subsection, based on the nonlinear model which can basically describe the fundamental dynamics properties of the wet limestone FGD systems, several linear state space models on different nominal operating points are identified so that a fuzzy switching law is raised to realize the control of the nonlinear system.

First, the conception of nonlinearity is introduced to weigh the disparities among different linear state space models. It is assumed that two finite-dimensional linear state models with the same input and output  $P_i(s)$ , ( $i = 1, 2$ ), whose left coprime factorization is  $P_i = N_i M^{-1}$ , ( $i = 1, 2$ ), then the gap metrics which measure nonlinearity of the two systems can be calculated as:

$$\delta(P_1, P_2) = \max(\vec{\delta}(P_1, P_2), \vec{\delta}(P_2, P_1)) \quad (17)$$

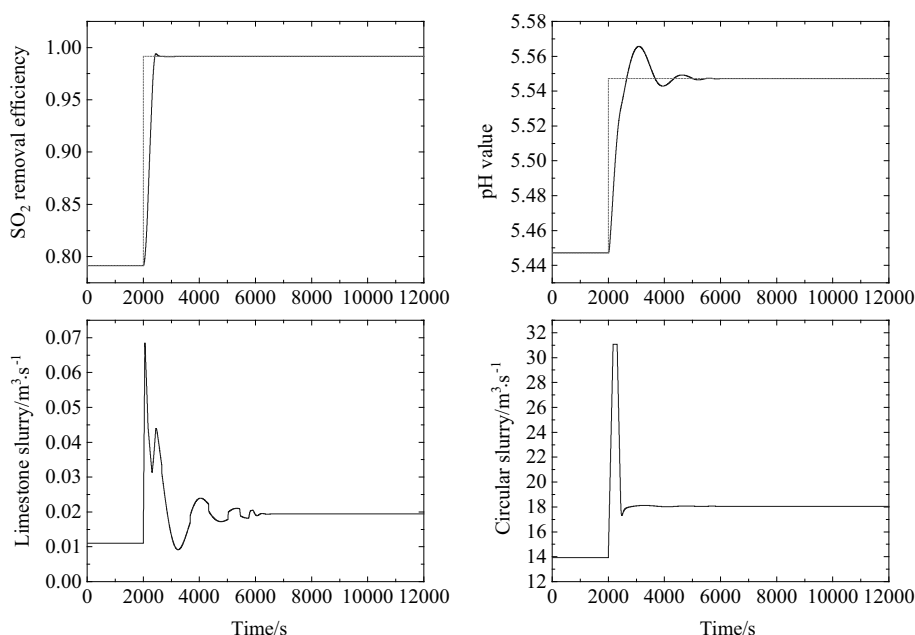


**Fig. 8.** Simulation results on the linear model under the LMPC control structure.

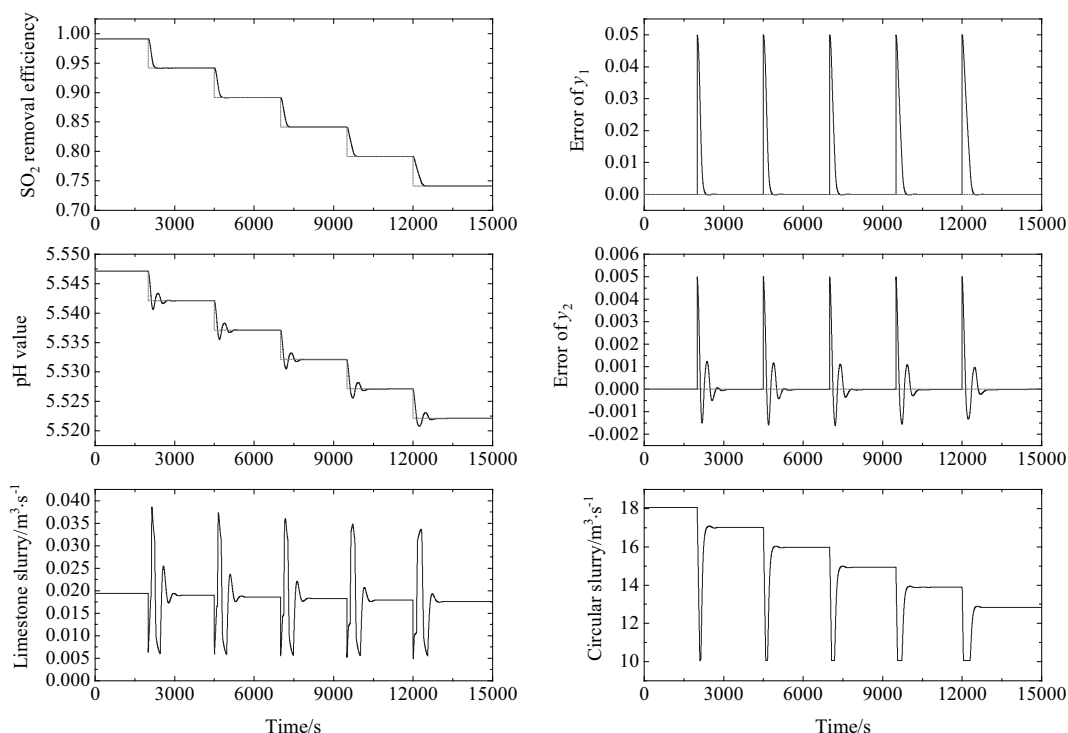


Fig. 9. The wide-range simulation results on the linear model under the LMPC control structure.

$$\vec{\delta}(P_i, P_j) = \inf_{Q \in H_{\infty}} (M_i M_j) - (M_j M_i) Q_{\infty}, i, j \in \{1, 2\}, i \neq j \quad (18)$$

Nonlinearity based on the usage of differential gap theory was adopted to find three nominal operating points by minimizing the value of the differential gap criterion. One typical operating condition, where  $y_1 = 99.2\%$ ,  $y_2 = 5.641$ ,  $u_1 = 0.0227 \text{ m}^3 \cdot \text{s}^{-1}$ ,  $u_2 = 25.4338 \text{ m}^3 \cdot \text{s}^{-1}$ , is chosen as the reference state space model. The value of differential gap between the reference model and other typical operating conditions is shown in Table 3.

According to the multimodal theory, a single linear controller cannot be used to control a whole nonlinear system if the differential gap among different operating conditions is too large to be ignored. In this case, more than one linear controllers should be used to eliminate control deviation caused by high nonlinearity. As is shown in Table 4, the largest gap among reference model and other typical operating conditions is 0.0045 so that the nonlinearity of system cannot be neglected. In the next subsection, a multimodality method will be derived to achieve set-point tracking in different operating conditions.

### 3.3.2. NMPC controller based on multimodality

In this subsection, a multimodality control scheme is derived based on the NMPC control algorithm (Fig. 10). It adopts several LMPC controllers together to control the system. To realize the structure, some critical

parameters reflecting nonlinearity during different operation conditions are put into the controllers. Then, the fuzzy law which designs the switching weight of different controllers is introduced to realize the NMPC scheme of the wet limestone FGD system (Fig. 11).

### 3.3.3. Analysis based on simulation results

The simulation results are shown in Figs. 12 and 13. It can be shown that the NMPC strategy tracks two different set points well. What's more, it can track the set point when the set points change largely. Until now, we obtained an applicable NMPC strategy for the wet limestone FGD system that can progressively track arbitrary set points well. This method will be used to make some economic improvements based on the multi-objective optimization, which is introduced in the next section.

## 4. Multi-objective Optimization NMPC

The classical MPC strategy is based on quadratic cost function whose target is to minimize the deviation of measured state values and values from the given set point. This set point is usually chosen as an optimal operating point in a steady state according to the field experience and actual demands. The set point can be adjusted in time so that the control system can perform well on energy saving, especially when the operating condition of system is under frequent changes for industrial systems with more frequent working conditions.

Table 3  
Seven typical operating conditions

| Operating condition | $u_1/\text{m}^3 \cdot \text{s}^{-1}$ | $u_2/\text{m}^3 \cdot \text{s}^{-1}$ | $y_1$  | $y_2$ |
|---------------------|--------------------------------------|--------------------------------------|--------|-------|
| A (52.5%)           | 0.0143                               | 6.6534                               | 0.5988 | 5.408 |
| B (60%)             | 0.0148                               | 8.6547                               | 0.6504 | 5.446 |
| C (70%)             | 0.0154                               | 12.9833                              | 0.7831 | 5.492 |
| D (75%)             | 0.0194                               | 14.2456                              | 0.7935 | 5.545 |
| E (80%)             | 0.0201                               | 14.7856                              | 0.8427 | 5.567 |
| F (90%)             | 0.0212                               | 18.3432                              | 0.9025 | 5.604 |
| G (100%)            | 0.0227                               | 25.4883                              | 0.992  | 5.641 |

Table 4  
Nonlinearity among seven typical operating conditions

|   | 1      | 2      | 3      | 4      | 5      | 6      | 7      |
|---|--------|--------|--------|--------|--------|--------|--------|
| 1 | 0.0000 | 0.0012 | 0.0025 | 0.0030 | 0.0033 | 0.0041 | 0.0045 |
| 2 | 0.0012 | 0.0000 | 0.0013 | 0.0018 | 0.0022 | 0.0029 | 0.0033 |
| 3 | 0.0025 | 0.0013 | 0.0000 | 0.0005 | 0.0009 | 0.0016 | 0.0020 |
| 4 | 0.0030 | 0.0018 | 0.0005 | 0.0000 | 0.0004 | 0.0011 | 0.0015 |
| 5 | 0.0033 | 0.0022 | 0.0009 | 0.0004 | 0.0000 | 0.0007 | 0.0011 |
| 6 | 0.0041 | 0.0029 | 0.0016 | 0.0011 | 0.0007 | 0.0000 | 0.0004 |
| 7 | 0.0045 | 0.0033 | 0.0020 | 0.0015 | 0.0011 | 0.0004 | 0.0000 |

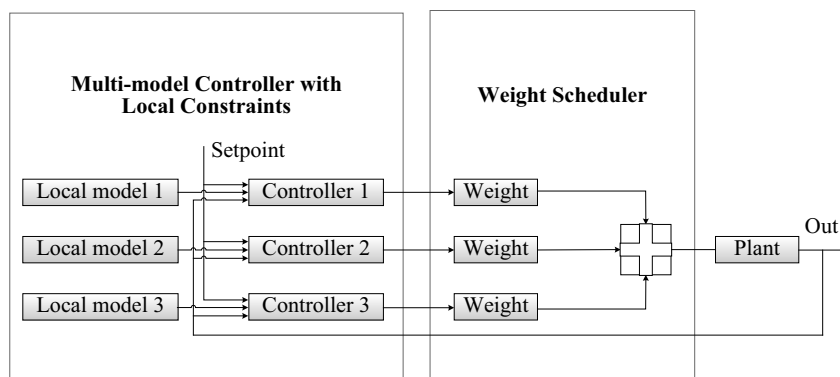


Fig. 10. Multimodality based NMPC control system.

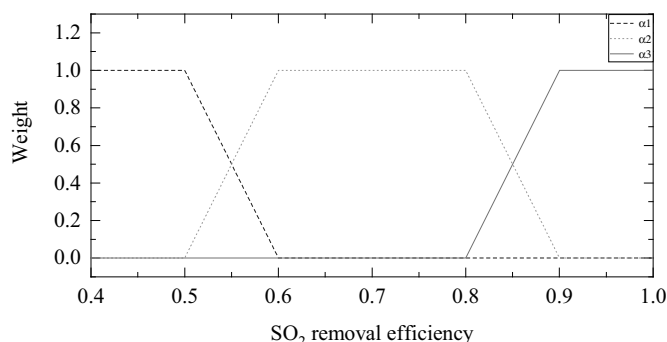


Fig. 11. Weighting function of gain scheduling.

#### 4.1. Problems on multi-objective optimization of NMPC scheme

The objective of optimal control is to maintain the outgoing  $\text{SO}_2$  concentration at selected set-points and to minimize the energy

consumption of pumps by manipulating the two control variables, the flow of the circular slurry pumps and the flow of the slurry pumps.

Among control parameters evaluated in economic performance, pH range > limestone reactivity and  $\text{SO}_2$  concentration > efficiency of particulate control devices > water re-circulation to the scrubber > entrainment of particles by the OUT-FGD gas, are the key factors of wet limestone FGD systems [40]. The pH of slurry tank is the most important process value which estimates the status of the wet limestone FGD system because it can influence the outgoing concentration of sulfur dioxide and the energy consumption of the process. Therefore, the optimization of the pH set point is an effective means to achieve the economic optimization of the wet limestone FGD system [41]. As a result, a multi-objective optimization control method is introduced to limit energy consumption to a reasonable range for energy saving. Based on the analysis of dynamics properties in the wet limestone FGD system, a multi-objective optimization algorithm, which concerns energy consumption and outgoing  $\text{SO}_2$  concentration, is constructed. The optimal set point is selected by simulation results under different

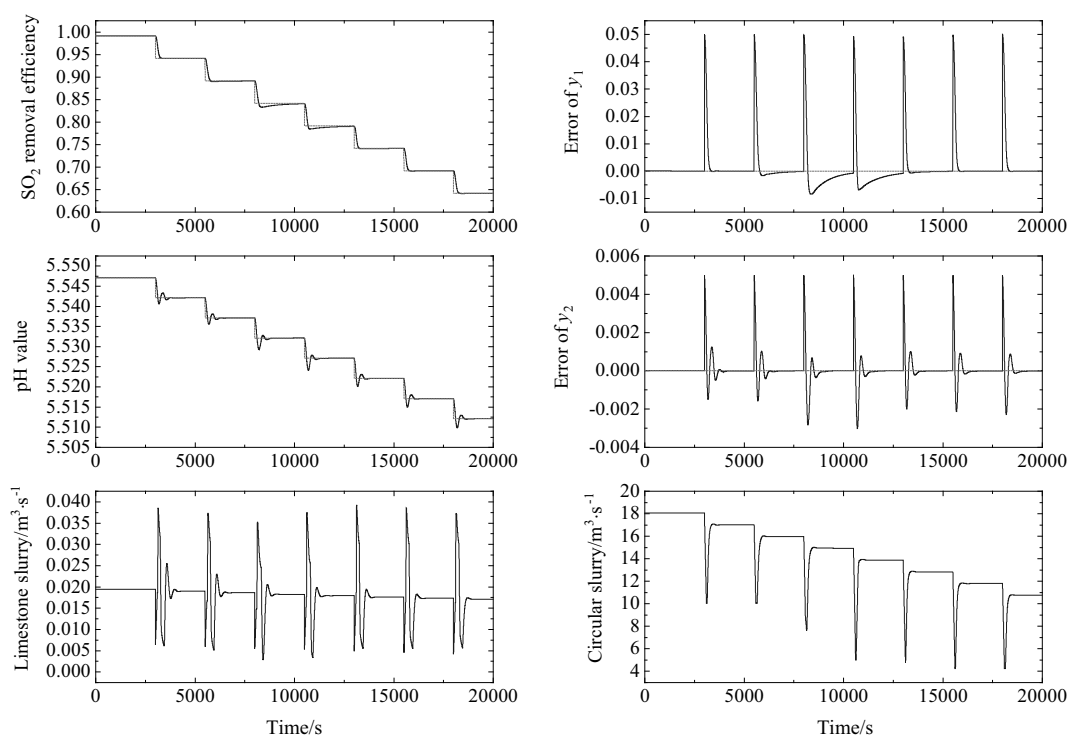


Fig. 12. The wide-range simulation results on the nonlinear model under the NMPC control structure.

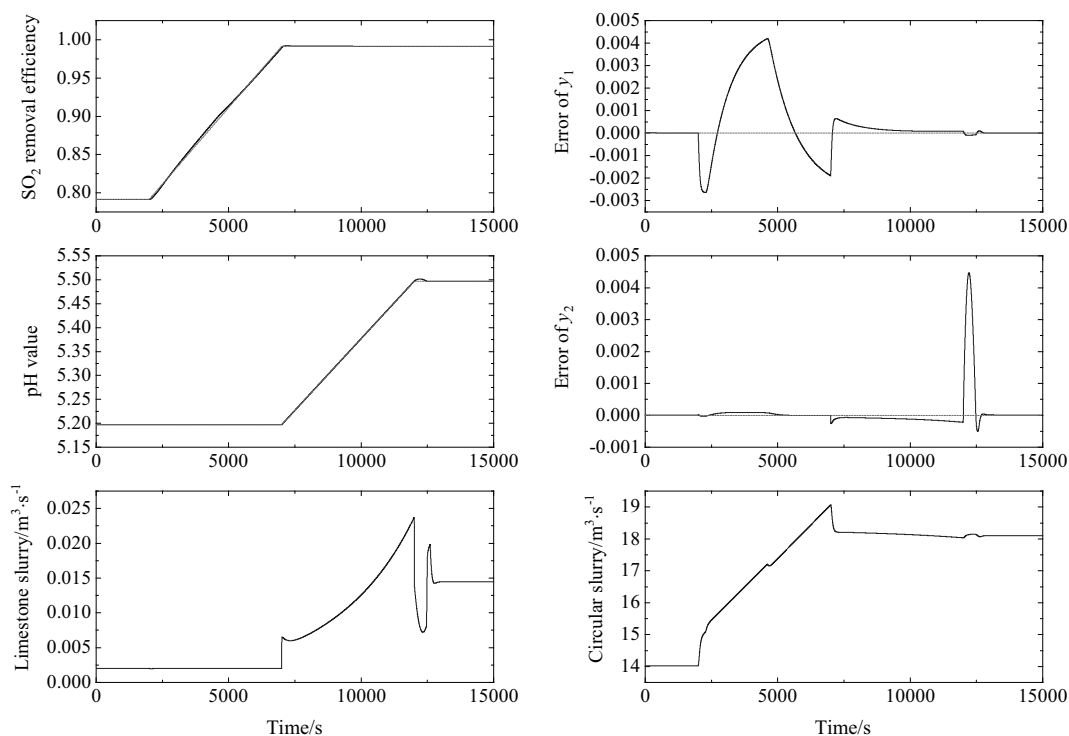


Fig. 13. Simulation results in a ramp signal on the nonlinear model under the NMPC control structure.

gas volume conditions so that the optimal set point of the pH of the slurry tank is obtained [42–45]. Then, the NMPC controller based on multimodality is employed to track the set points of outgoing concentration of sulfur dioxide regulated by environmental policy and the optimized pH value simultaneously [46].

The multi-objective optimization of NMPC control scheme is introduced in Fig. 14. It is shown in Fig. 14 that the optimization solving contains the following steps. Firstly, the prediction model predicts the future behavior of the process within a predetermined time interval (namely within the prediction horizon) based on the optimal pH value. Secondly, a series of controlled variables are obtained by minimizing the objective function within another time interval (namely within the control horizon). Thirdly, the first values of the control variables are applied to the WFGD system. Based on the process above, the optimization solving is accomplished.

## 4.2. Simulation results

### 4.2.1. NMPC performance of pH optimization

As is shown in Fig. 15, lower pH value can improve the economic performance of system. By setting the pH value around 5.35, not only can the set point of  $\text{SO}_2$  removal efficiency be tracked well but also the economic objective can be satisfied. However the set point has a lower limit to ensure the basic control objective, the set point tracking of  $\text{SO}_2$  removal efficiency because the capability of  $\text{SO}_2$  absorption will be weakened as the pH value decreases.

For more detailed exposition, an illustrative example is given where the average flue gas volume is set to  $735 \text{ m}^3 \cdot \text{s}^{-1}$  and the lower control limit of  $\text{SO}_2$  removal efficiency is set to 98%. As is shown in Fig. 16, the  $\text{SO}_2$  removal efficiency increases as the pH value increases. When the pH value is in the range from 5.2 to 5.438, the economic cost is in a

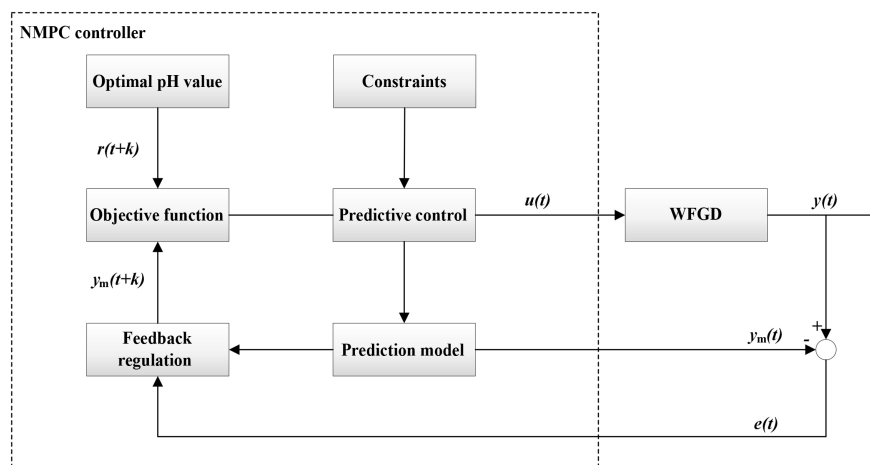


Fig. 14. NMPC scheme with multi-objective optimization.

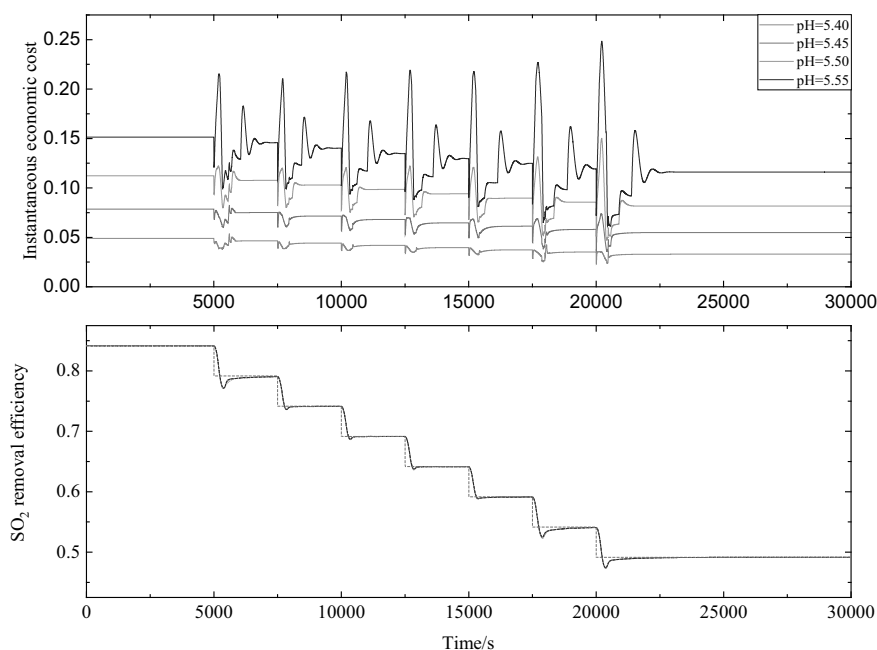


Fig. 15. The wide-range simulation results under the NMPC control structure containing optimization of instantaneous economic cost.

lower level but the control system cannot fulfill the lower control limit of  $\text{SO}_2$  removal efficiency. When the pH value reaches 5.438, the wet limestone system is in a critical state where consumes lowest energy and fulfills the lower control limit of  $\text{SO}_2$  removal efficiency.

#### 4.2.2. NMPC performance of multi-objective optimization

The simulation is set as follows: the conventional control objective is to ensure  $\text{SO}_2$  removal efficiency to be no less than the lower limit, the economic objective is minimizing the operational electricity consumption cost, the measurable disturbance is boiler load. Boiler load fluctuates between 700 MW to 1000 MW. Meanwhile, the lower limit of emission index is 98%. The sampling time is set to be 1 s, and the prediction horizon and control horizon are both set to be 100 s.

Fig. 17 shows the dynamic curves of control variables and economic cost under boiler load fluctuation. It is shown that NMPC scheme minimize operational electricity consumption cost and track emission

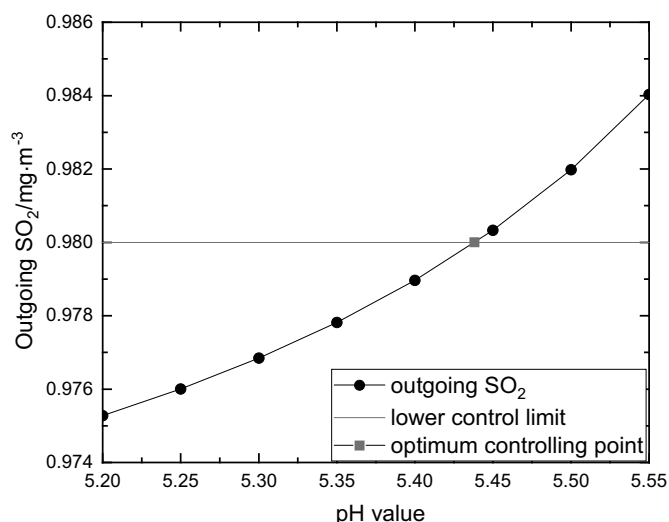


Fig. 16. The relation curve of  $\text{SO}_2$  removal efficiency with the pH value when the average flue gas volume is  $735 \text{ m}^3 \cdot \text{s}^{-1}$ .

performance index under large variation of boiler load by introducing optimal pH value and set point into the design.

## 5. Conclusions

The purpose of this paper is to realize the multi-objective optimization control of the wet limestone FGD system based on NMPC strategy. With reasonable simplifications, a nonlinear mathematical model was proposed and verified by field data. Based on the derived model, the prediction model of NMPC was obtained by using multimodality strategy. To improve the economic performance, a multi-objective optimization of NMPC control strategy was proposed. According to the performance of the derived control strategy, the advantages of the NMPC was confirmed theoretically in terms of the following aspects, a) Control deviations and mismatch in conventional scheme caused by feedforward signals which are difficult to determine, e.g., the flue gas volume signal, are lighten by the prediction model in NMPC scheme, e.g., the steady-state errors of  $\text{SO}_2$  removal efficiency and pH value are weakened to zero; b) The multi-objective optimization requirements of the wet limestone FGD system can be satisfied by the NMPC scheme, e.g., set point tracking and energy saving; c) The economic performance of the system can be improved based on the control framework, e.g., the economic cost is increased by 1.58% in variable condition.

Along the same research line, the work in the future will be paid attention to introducing critical parameters in boiler side into the NMPC frame to further improve the control performance.

## Acknowledgments

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## Appendix A

$$N_{\text{SO}_2, A} = K_{G, \text{SO}_2} \cdot \alpha_A \cdot (p_{\text{SO}_2} - H_{\text{SO}_2} \cdot C_{\text{SO}_2}) \quad (\text{A1})$$

$$N_{\text{CO}_2, A} = k_{L, \text{CO}_2} \cdot \alpha_A \cdot \left( \frac{p_{\text{CO}_2}}{H_{\text{CO}_2}} - C_{\text{CO}_2} \right) \quad (\text{A2})$$

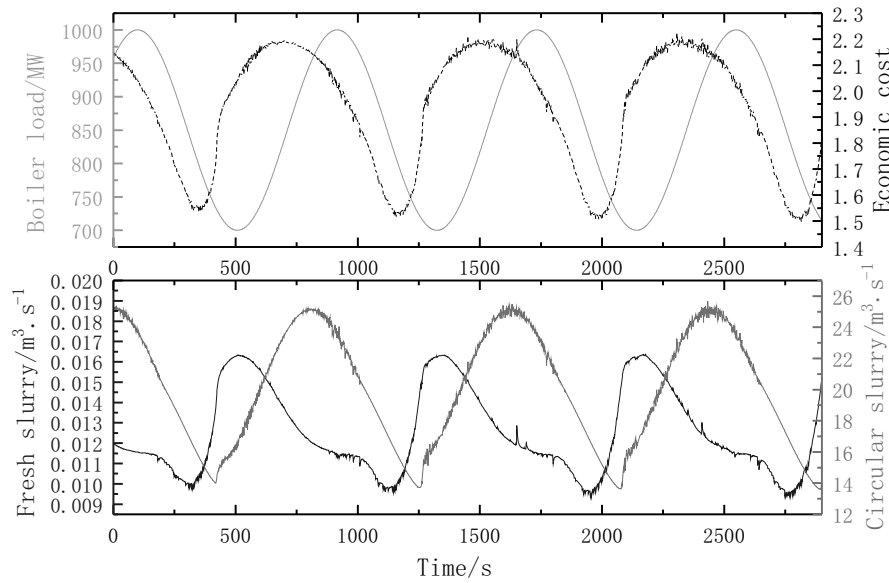


Fig. 17. Simulation results of multi-objective optimization under the NMPC structure.

$$p_{\text{SO}_2} = n_{\text{SO}_2} \cdot R \cdot T / G \quad (\text{A3})$$

$$N_{\text{SO}_2, \text{T}} = K_{\text{G, SO}_2} \cdot \alpha_{\text{T}} \cdot (p_{\text{SO}_2} - H_{\text{SO}_2} \cdot C_{\text{SO}_2}) \quad (\text{A4})$$

$$N_{\text{CO}_2, \text{T}} = E_{\text{CO}_2} \cdot k_{\text{L, CO}_2} \cdot \alpha_{\text{T}} \cdot \left( \frac{p_{\text{CO}_2}}{H_{\text{CO}_2}} - C_{\text{CO}_2} \right) \quad (\text{A5})$$

where  $N_{\text{SO}_2, \text{A}}$ ,  $N_{\text{CO}_2, \text{A}}$  is the absorption rate of  $\text{SO}_2$ ,  $\text{CO}_2$  in the scrubber,  $N_{\text{SO}_2, \text{T}}$ ,  $N_{\text{CO}_2, \text{T}}$  is the absorption rate of  $\text{SO}_2$ ,  $\text{CO}_2$  in the slurry tank,  $p_{\text{SO}_2}$ ,  $p_{\text{CO}_2}$  is the partial pressure of  $\text{SO}_2$ ,  $\text{CO}_2$ ,  $H_{\text{SO}_2}$ ,  $H_{\text{CO}_2}$  is the Henry coefficient of  $\text{SO}_2$ ,  $\text{CO}_2$ ,  $C_{\text{SO}_2}$ ,  $C_{\text{CO}_2}$  is the concentration of  $\text{SO}_2$ ,  $\text{CO}_2$  in liquid phase,  $K_{\text{L, SO}_2}$  is mass transfer coefficient of  $\text{SO}_2$  in liquid phase,  $K_{\text{G, SO}_2}$  is the total mass transfer coefficient of  $\text{SO}_2$ ,  $\alpha_{\text{T}}$  is the gas-liquid specific surface area in the scrubber,  $\alpha_{\text{T}}$  is the gas-liquid specific surface area in the slurry tank,  $n_{\text{SO}_2}$  is the molar flow of  $\text{SO}_2$  in raw flue gas,  $G$  is the average flow of the flue gas,  $E_{\text{CO}_2}$  is the activation energy of  $\text{CO}_2$ .

$$R_{\text{Sgy}} = \frac{C_{\text{Ca}^{2+}} \cdot C_{\text{SO}_4^{2-}}}{L_{\text{gy}}} \quad (\text{A6})$$

$$K_{\text{gy}} = 0.00011 \cdot A_{\text{gy}} \quad (\text{A7})$$

$$A_{\text{gy}} = \frac{C_{\text{gy}} \cdot M_{\text{gy}}}{\rho_{\text{gy}} \cdot \left( \frac{3}{4} \cdot \pi \cdot \left( \frac{d_{\text{gy}}}{2} \right)^3 \right)} \cdot \left( 4 \cdot \pi \cdot \left( \frac{d_{\text{gy}}}{2} \right)^2 \right) \quad (\text{A8})$$

where  $C_{\text{Ca}^{2+}}$ ,  $C_{\text{SO}_4^{2-}}$  is the concentration of  $\text{Ca}^{2+}$ ,  $\text{SO}_4^{2-}$ ,  $L_{\text{gy}}$  is the equilibrium solution constant of gypsum,  $A_{\text{gy}}$  is the surface area of gypsum particles per unit volume,  $M_{\text{gy}}$  is the molar mass of gypsum,  $d_{\text{gy}}$  is the average diameter of gypsum.

$$\Delta(C_{\text{CO}_2} + C_{\text{HCO}_3^-} + C_{\text{CO}_3^{2-}}) = \tau_{\text{A}} \cdot (N_{\text{CO}_2, \text{A}} - R_{\text{CaCO}_3}) \quad (\text{A9})$$

$$\Delta(C_{\text{gy}}) = \tau_{\text{A}} \cdot R_{\text{gy}} \quad (\text{A10})$$

$$\Delta(C_{\text{SO}_4^{2-}} + C_{\text{HSO}_4^-}) = \tau_{\text{A}} \cdot (R_{\text{ox}} - R_{\text{gy}}) \quad (\text{A11})$$

$$\Delta(C_{\text{HCO}_3^-} + C_{\text{CO}_3^{2-}}) = \tau_{\text{A}} \cdot (R_{\text{CaCO}_3}) \quad (\text{A12})$$

$$\Delta(C_{\text{Ca}^{2+}}) = \tau_{\text{A}} \cdot (R_{\text{CaCO}_3} - R_{\text{gy}}) \quad (\text{A13})$$

$$\Delta(C_{\text{CaCO}_3}) = -\tau_{\text{A}} \cdot R_{\text{CaCO}_3} \quad (\text{A14})$$

Table A1

Partial differential and algebraic equations of dynamic characteristics of the gas-to liquid contract zone

| No. | Partial differential and algebraic equations                                                                                                                                                                                                           |
|-----|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 1   | $\partial(C_{\text{SO}_2} + C_{\text{HSO}_3^-} + C_{\text{SO}_3^{2-}})/\partial t = \tau_{\text{A}} \cdot (N_{\text{SO}_2, \text{A}} - R_{\text{ox}})$                                                                                                 |
| 2   | $\partial(C_{\text{CO}_2} + C_{\text{HCO}_3^-} + C_{\text{CO}_3^{2-}})/\partial t = \tau_{\text{A}} \cdot (N_{\text{CO}_2, \text{A}} - R_{\text{CaCO}_3})$                                                                                             |
| 3   | $\partial(C_{\text{gy}})/\partial t = \tau_{\text{A}} \cdot R_{\text{gy}}$                                                                                                                                                                             |
| 4   | $\partial(C_{\text{SO}_4^{2-}} + C_{\text{HSO}_4^-})/\partial t = \tau_{\text{A}} \cdot (R_{\text{ox}} - R_{\text{gy}})$                                                                                                                               |
| 5   | $\partial(C_{\text{HCO}_3^-} + C_{\text{CO}_3^{2-}})/\partial t = \tau_{\text{A}} \cdot (R_{\text{CaCO}_3})$                                                                                                                                           |
| 6   | $\partial(C_{\text{Ca}^{2+}})/\partial t = \tau_{\text{A}} \cdot (R_{\text{CaCO}_3} - R_{\text{gy}})$                                                                                                                                                  |
| 7   | $\partial(C_{\text{CaCO}_3})/\partial t = -\tau_{\text{A}} \cdot R_{\text{CaCO}_3}$                                                                                                                                                                    |
| 8   | $N_{\text{SO}_2, \text{A}} = K_{\text{G, SO}_2} \cdot \alpha_{\text{A}} \cdot (p_{\text{SO}_2} - H_{\text{SO}_2} \cdot C_{\text{SO}_2})$                                                                                                               |
| 9   | $N_{\text{CO}_2, \text{A}} = k_{\text{L, CO}_2} \cdot \alpha_{\text{A}} \cdot \left( \frac{p_{\text{CO}_2}}{H_{\text{CO}_2}} - C_{\text{CO}_2} \right)$                                                                                                |
| 10  | $p_{\text{SO}_2} = n_{\text{SO}_2} \cdot R \cdot T / G$                                                                                                                                                                                                |
| 11  | $N_{\text{SO}_2, \text{T}} = K_{\text{G, SO}_2} \cdot \alpha_{\text{T}} \cdot (p_{\text{SO}_2} - H_{\text{SO}_2} \cdot C_{\text{SO}_2})$                                                                                                               |
| 12  | $N_{\text{CO}_2, \text{T}} = E_{\text{CO}_2} \cdot k_{\text{L, CO}_2} \cdot \alpha_{\text{T}} \cdot \left( \frac{p_{\text{CO}_2}}{H_{\text{CO}_2}} - C_{\text{CO}_2} \right)$                                                                          |
| 13  | $R_{\text{Sgy}} = \frac{C_{\text{Ca}^{2+}} \cdot C_{\text{SO}_4^{2-}}}{L_{\text{gy}}}$                                                                                                                                                                 |
| 14  | $\frac{dC_{\text{Ca}^{2+}}}{dt} = T_{\text{Ca}^{2+}} + R_{\text{CaCO}_3} - R_{\text{gy}}$                                                                                                                                                              |
| 15  | $K_{\text{gy}} = 0.00011 \cdot A_{\text{gy}}$                                                                                                                                                                                                          |
| 16  | $A_{\text{gy}} = \frac{C_{\text{gy}} \cdot M_{\text{gy}}}{\rho_{\text{gy}} \cdot \left( \frac{3}{4} \cdot \pi \cdot \left( \frac{d_{\text{gy}}}{2} \right)^3 \right)} \cdot \left( 4 \cdot \pi \cdot \left( \frac{d_{\text{gy}}}{2} \right)^2 \right)$ |
| 17  | $R_{\text{ox}} = K_{\text{ox}} \cdot (C_{\text{HSO}_3^-} + C_{\text{SO}_3^{2-}})^{1.5}$                                                                                                                                                                |
| 18  | $R_{\text{CaCO}_3} = k_{\text{CaCO}_3} \cdot (1 - R_{\text{Sgy}})$                                                                                                                                                                                     |
| 19  | $R_{\text{Sgy}} = \frac{C_{\text{Ca}^{2+}} \cdot C_{\text{CO}_3^{2-}}}{K_{\text{CP}}}$                                                                                                                                                                 |
| 20  | $R_{\text{gy}} = K_{\text{gy}} \cdot (R_{\text{Sgy}} - 1)$                                                                                                                                                                                             |

Table A2

Difference equations of dynamic characteristics of the gas-to liquid contract zone

| No. | Difference equations                                                                                                                          |
|-----|-----------------------------------------------------------------------------------------------------------------------------------------------|
| 1   | $\Delta(C_{\text{SO}_2} + C_{\text{HSO}_3^-} + C_{\text{SO}_3^{2-}}) = \tau_{\text{A}} \cdot (N_{\text{SO}_2, \text{A}} - R_{\text{ox}})$     |
| 2   | $\Delta(C_{\text{CO}_2} + C_{\text{HCO}_3^-} + C_{\text{CO}_3^{2-}}) = \tau_{\text{A}} \cdot (N_{\text{CO}_2, \text{A}} - R_{\text{CaCO}_3})$ |
| 3   | $\Delta(C_{\text{gy}}) = \tau_{\text{A}} \cdot R_{\text{gy}}$                                                                                 |
| 4   | $\Delta(C_{\text{SO}_4^{2-}} + C_{\text{HSO}_4^-}) = \tau_{\text{A}} \cdot (R_{\text{ox}} - R_{\text{gy}})$                                   |
| 5   | $\Delta(C_{\text{HCO}_3^-} + C_{\text{CO}_3^{2-}}) = \tau_{\text{A}} \cdot (R_{\text{CaCO}_3})$                                               |

Table A2 (continued)

| No. | Difference equations                                       |
|-----|------------------------------------------------------------|
| 6   | $\Delta(C_{Ca^{2+}}) = \tau_A \cdot (R_{CaCO_3} - R_{gy})$ |
| 7   | $\Delta(C_{CaCO_3}) = -\tau_A \cdot R_{CaCO_3}$            |

## References

- [1] D. Wu, X.Z. Ma, S.Q. Zhang, Integrating synergistic effects of air pollution control technologies: More cost-effective approach in the coal-fired sector in China, *J. Clean. Production* 199 (20) (2018) 1035–1042.
- [2] C.L. Gage, Limestone dissolution in modelling of slurry scrubbing for flue gas desulfurization, Ph.D. Thesis, University of Texas, Texas 1989.
- [3] R.S. Argarwal, G.T. Rochelle, Chemistry of Limestone Slurry Scrubbing, SO<sub>2</sub> Control Symposium 3, 2015.
- [4] C. Brogren, H.T. Karlson, A model for prediction of limestone dissolution in wet flue gas desulfurization applications, *Ind. Eng. Chem. Res.* 36 (9) (1997) 3889–3897.
- [5] D. Eden, M. Luckas, A heat and mass transfer model for the simulation of the wet limestone flue gas scrubbing process, *Chem. Eng. Technol.* 21 (1) (1998) 56–60.
- [6] H. Kikkawa, H. Kaku, S. Nozawa, K. Kohata, New wet FGD process using granular limestone, *Proceedings of power gen Europe* (1997) 585–601.
- [7] J. Warych, M. Szymanowski, Model of the wet limestone flue gas desulfurization process for cost optimization, *Ind. Eng. Chem. Res.* 40 (12) (2001) 2597–2605.
- [8] S. Kiil, M.L. Michelsen, K.D. Johansen, Experimental investigation and modelling of a wet flue gas desulphurisation pilot plant, *Ind. Eng. Chem. Res.* 37 (1998) 2792–2806.
- [9] M.X. Guo, Studies of slurry jet flue gas desulfurization, Ph. D. Thesis, Tsinghua Univ., Beijing, (2002).
- [10] L. Wurth, R. Hannemann, W. Marquardt, Neighboring-extremal updates for nonlinear model-predictive control and dynamic real-time optimization, *J. Process Control* 19 (8) (2009) 1277–1288.
- [11] H.G. Bock, M.M. Diehl, D.B. Leineweber, J.P. Schlöder, A direct multiple shooting method for real-time optimization of nonlinear DAE processes, *J. Process Control* 26 (2009) 245–267.
- [12] F. Allgöwer, A. Zheng, Nonlinear model predictive control, progress in systems and control theory, *Birkhäuser Verlag* 26 (2000) 245–267.
- [13] M. Diehl, H. Bock, J. Schlöder, R. Findeisen, Z. Nagy, F. Allgöwer, Real-time optimization and nonlinear model predictive control of processes governed by differential-algebraic equations, *J. Process Control* 12 (4) (2002) 577–585.
- [14] L. Grüne, J. Pannek, Analysis of unconstrained NMPC schemes with incomplete optimization, *IFAC Proceedings* 43 (14) (2010) 238–243.
- [15] V.M. Zavala, M. Anitescu, Real-time nonlinear optimization as a generalized equation, *SIAM J. Control. Optim.* 48 (2010) 5444–5467.
- [16] I.J. Wolf, L. Würth, W. Marquardt, Rigorous solution vs. fast update: acceptable computational delay in NMPC, 50th IEEE CDC-ECC 2011, pp. 5230–5235.
- [17] V.M. Zavala, L.T. Biegler, The advanced-step NMPC controller: optimality, stability and robustness, *Automatica* 45 (1) (2009) 86–93.
- [18] J. Jäschke, X. Yang, L.T. Biegler, Fast economic model predictive control based on NLP-sensitivities, *J. Process Control* 24 (8) (2014) 1260–1272.
- [19] M.V. Sotnikova, Control system design for visual positioning of a ship based on NMPC and multi-objective structure, *IFAC-PapersOnLine* 51 (32) (2018) 445–450.
- [20] O. Makoto, T. Gaku, Real-time autonomous car motion planning using NMPC with approximated problem considering traffic environment, *IFAC-PapersOnLine* 51 (20) (2018) 279–286.
- [21] W.Y. Zhang, D.X. Huang, Y.D. Wang, J.C. Wang, Adaptive state feedback predictive control and expert control for a delayed coking furnace, *Chin. J. Chem. Eng.* 16 (4) (2008) 590–598.
- [22] H. Flemming, M. Alexander, T.B. Lorenz, Multistage NMPC with on-line generated scenario trees: Application to a semi-batch polymerization process, *J. Process Control* 80 (2019) 167–179.
- [23] M.Z. Yu, T.B. Lorenz, Economic NMPC strategies for solid sorbent-based CO<sub>2</sub> capture, *IFAC-PapersOnLine* 51 (18) (2018) 103–108.
- [24] D. Telen, B. Houska, M. Vallerio, F. Logist, J. Vanimpe, A study of integrated experiment design for NMPC applied to the Droop model, *Chem. Eng. Sci.* 160 (2017) 370–383.
- [25] A.V. Fiacco, G.P. McCormick, Nonlinear Programming: Sequential Unconstrained Minimization Techniques, John Wiley & Sons Inc., 1968.
- [26] S.H. Chen, H.M. He, X.F. Li, G. Chen, Application of fuzzy PID controller with self-adaptive algorithm and non-uniform grid scheduling to WFGD, *IFAC Proceedings Volumes* 42 (9) (2009) 20–25.
- [27] A. Gautam, Y.C. Chu, Y.C. Soh, Optimized dynamic policy for receding horizon control of linear time-varying systems with bounded disturbances, *IEEE Trans Automat Control* 57 (4) (2012) 973–988.
- [28] Alexandra Grancharova, Tor A. Johansen, Computation, approximation and stability of explicit feedback min–max nonlinear model predictive control, *Automatica* 45 (5) (2009) 1134–1143.
- [29] D.F. Hen, H. Huang, Q.X. Chen, Quasi-min–max MPC for constrained nonlinear systems with guaranteed input-to-state stability, *Journal of the Franklin Institute* 351 (6) (2014) 3405–3423.
- [30] Y. Zhong, X. Gao, W. Huo, Z.Y. Luo, M.J. Ni, K.F. Cen, A model for performance optimization of wet flue gas desulfurization systems of power plants, *Fuel Process. Technol.* 89 (11) (2008) 1025–1032.
- [31] G. Helle, K. Søren, E.J. Jan, N.J. Jørgen, H. Jørn, F. Folmer, D.J. Kim, Full-scale measurements of SO<sub>2</sub> gas phase concentrations and slurry compositions in a wet flue gas desulphurisation spray absorber, *Fuel* 83 (9) (2004) 1151–1164.
- [32] Q. Zhang, S.J. Wang, P. Zhu, Z.Y. Wang, G. Zhang, Full-scale simulation of flow field in ammonia-based wet flue gas desulfurization double tower, *J. Energy Inst.* 91 (4) (2018) 619–629.
- [33] C. Patricia, Status of flue gas desulphurisation (FGD) systems from coal-fired power plants: Overview of the physico-chemical control processes of wet limestone FGDs, *Fuel* 144 (2015) 274–286.
- [34] X.H. Li, Y.D. He, Simulation of flue gas flow field in the spraying tower of wet flue gas desulfurization system Therm, *Power Generation Technologies* 39 (2010) 41–45.
- [35] L.Q. Li, C. Zhang, G.J. Huang, Z. Liu, W.W. Ma, A multicomponent droplet model in simulating mass transfer of the ammonia-based spray process, *Proc. CSEE* 34 (2014) 5741–5749.
- [36] S.L. Zhao, Y.F. Duan, J.C. Lu, R. Gupt, D. Pudasainee, S. Liu, M. Liu, J.H. Lua, Thermal stability, chemical speciation and leaching characteristics of hazardous trace elements in FGD gypsum from coal-fired power plants, *Fuel* 231 (2018) 94–100.
- [37] P. Córdoba, R. Ochoa-Gonzalez, O. Font, M. Izquierdo, X. Querol, C. Leiva, Partitioning of trace inorganic elements in a coal-fired power plant equipped with a wet flue gas desulphurisation system, *Fuel* 92 (2012) 145–157.
- [38] R. Meij, T.H. Winkel, The emissions and environmental impact of PM10 and trace elements from a modern coal-fired power plant equipped with ESP and wet FGD, *Fuel Process. Technol.* 85 (2004) 641–656.
- [39] C. Claudio, D.B. Cataldo, M. Marianna, P. Raffaele, W. Tapio, Ultrasonic enhanced limestone dissolution: Experimental and mathematical modeling, *Chemical Engineering and Processing: Process Intensification* 118 (2017) 26–36.
- [40] J. Rawlings, D. Mayne, Model Predictive Control: Theory and Design, Nob Hill Publishing, San Francisco, 2009.
- [41] G. Lars, NMPC without terminal constraints, *IFAC Proceedings* 45 (17) (2012) 1–13.
- [42] A.W. Hermansson, S. Syafie, An offset-free MPC formulation for nonlinear systems using adaptive integral controller, *ISA Trans.* 91 (2019) 66–77.
- [43] B.W. Frandsen, K. Søren, E.J. Jan, Optimisation of a wet FGD pilot plant using fine limestone and organic acids, *Chem. Eng. Sci.* 56 (10) (2001) 3275–3287.
- [44] Z. Mario, G. Sébastien, D. Moritz, A tracking MPC formulation that is locally equivalent to economic MPC, *J. Process Control* 45 (2016) 30–42.
- [45] M. Ellis, P.D. Christofides, Economic model predictive control with time-varying objective function for nonlinear process systems, *AIChE J.* 60 (2014) 507–519.
- [46] M. Zanon, S. Gros, M. Diehl, Indefinite linear MPC and approximated economic MPC for nonlinear systems, *J. Process Control* 24 (2014) 1273–1281.