Eulerian–Eulerian Numerical Study of the Flue Gas Desulfurization Process in a Semidry Spouted Bed Reactor

Feng Wu,* Jinhao Bai, Kai Yue, Ming Gong, Xiaoxun Ma, and Wenjing Zhou

ABSTRACT: The Eulerian–Eulerian two-fluid model (TFM) in conjunction with kinetic theory of granular flows (KTGF) was used for analyzing water vaporization and the semidry flue gas desulfurization process in a two-dimensional powder–particle spouted bed (PPSB). In an environment with high-temperature gas, desulfurization slurry is wrapped on the surface of moving particles and evaporated, along with the application of the user defined function (UDF) method to accomplish water heat and mass transfer by considering evaporation in the simulation process. The simulation results revealed that the best mass- and heat-transfer effect of each phase can be found in the outer annulus and the near spout region, both of which are also the main areas where water vaporization occurs. The rate of desulfurization products decreases with the increase in inlet gas temperature as the water vaporization rate increases. The volume fraction of desulfurization reaction products decreases with the increase in inlet flue gas temperature. Compared with other working conditions, the highest desulfurization efficiency reaches 84% when the inlet flue gas temperature is 480 K. The change of the desulfurization product rate with the radial distance is the same under different superficial gas velocities, with the peak desulfurization efficiency appearing in the annulus. The optimal operating parameter for the desulfurization process is available in PPSB, and the desulfurization efficiency and gas handling capacity reach the best result when the superficial gas velocity equals 1.2 Ums.

1. INTRODUCTION

It is well known that sulfur dioxide can cause a variety of hazards to the environment and human health. As far as the environment is concerned, one of the most destructive effects of sulfur dioxide is the so-called acid rain. More specifically, more and more fossil fuels are used to meet increasing energy demand, resulting in the increased atmospheric emission of sulfur dioxide and carbon dioxide, thus imposing a great danger to the environment and human health. The emission of SO2 and CO2 from power plants burning fossil fuels causes acid rain in the atmosphere, thereby resulting in serious environmental problems. Hence, the removal of SO2 and CO2 from gas has been a worldwide concern. Various technologies for flue gas desulfurization (FGD) can be classified into three different types, namely, wet scrubber, semidry processes, and dry processes. Mohanty et al. developed a three-stage countercurrent multistage fluidized-bed adsorption desulfurization device and carried out an experimental study on the removal of sulfur dioxide from flue gas based on a two-phase system under the condition of continuous bubbling fluidization. Zhang and Gui illustrated that high desulfurization efficiency can be achieved by a magnetic fluidized bed (MFB) as a reactor and adopting a semidry flue gas desulfurization (FGD) process. Tao et al. established a detailed model to simulate the process characterized by multistage humidified flue gas desulfurization in an underfeed circulating spouted bed (UCSB). Even more, they also developed a three-dimensional mathematical model to couple complex flow and chemical reactions, which can be conducive to system analysis and desulfurization data analysis. Used in industry, the semidry flue gas desulfurization process developed during the 1980s. Spray drying tower, as a typical semidry flue gas desulfurization process, can remove SO2 from flue gas effectively, thereby serving as a substitute for wet scrubber.

Several studies on spouted bed flue gas purification, such as the removal of sulfur dioxide and carbon dioxide, have been carried out. Ma et al. experimentally investigated the application of limestone for the removal of SO2 from flue gas in the semidry FGD process with a powder–particle spouted bed (PPSB), along with the exploration of the effect exerted by gas components on the removal of SO2 from flue gas in the semidry FGD process with a PPSB. However, without building a mathematical model of the desulfurization process, they analyzed it in a powder–particle spouted bed. Xu et al.
experimentally tested a new semidry desulfurization process in which the powder–particle spouted bed (PPSB) was used. Wu et al. investigated the influence exerted by the pore-size distribution of lime on the reactivity for the removal of SO2 in the presence of high-concentration CO2 at high temperatures. Wang and Zhang experimentally analyzed the effects exerted by humidification water parameters on the desulfurization removal efficiency of semidry flue gas. Also, a mathematical model for investigating the removal of SO2 and CO2 in a powder–particle spouted bed under a nonisothermal operating condition was developed by Rahimi et al. A laboratory-scale PPSB is employed by Haghmeaghadar et al. to investigate the effects exerted by operating parameters on the removal efficiency of CO2. Extensive numerical simulation of PPSB has been carried out, while only a few simulations were focused on mathematical modeling in a spouted bed reactor. Mathematical modeling of a one-dimensional isothermal model in spouting beds was first proposed by Mathur et al. Some mathematical models to analyze the performance capability of a spouted bed in different processes have been explored. Recently, Fakhari et al. experimentally analyzed the simultaneous absorption of CO2 and SO2 by a NaOH solution in a PPSB reactor and Niksia et al. established a one-dimensional mathematical model to predict the behavior of the spouted bed reactor in the gasification process of pistachio shell pyrolysis carbon. Tseng et al., on studying the interactions between the flow structure and chemical reaction, numerically simulated a flue gas desulfurization tower with perforated sieve trays by an Eulerian–Eulerian method, along with the implementation of the species transport equations of the chemical reactions in the study.

To our knowledge, no mathematical modeling has been reported to investigate the desulfurization process in PPSB. Besides, it is clear that the operating temperature and superficial velocity of the inlet gas impose a significant impact on the desulfurization efficiency of PPSB, making it necessary to analyze the effect mechanism of temperature and velocity on the desulfurization process. In this study, computational fluid dynamics (CFD) combined with a two-fluid model (TFM) was used for simulating water vaporization and a semidry flue gas desulfurization process in the PPSB on the basis of the gas–solid two-phase flow. The mathematical and physical models of the water vaporization process and flue gas desulfurization reaction process have been established through reasonable hypothesis and simplification of the system, along with the application of the numerical simulations to analyze the effects exerted by the operating temperature and superficial velocity on the desulfurization efficiency in the PPSB.

2. COMPUTATIONAL METHOD

2.1. Governing Equations. The adsorbent slurry is continuously fed into the spouted bed, in which the coarse particles are spouted by the hot gas containing sulfur dioxide. Then, the desulfurization slurry absorbs sulfur dioxide in the flue gas with the circulation of the particles and is evaporated by the high-temperature flue gas. The desulfurization product adheres to the surface of coarse particles after water is vaporized. Finally, the unused adsorbent particles and reaction products are entrained by clean gas in the form of dry powder and collected by a bag filter. In short, this section summarizes the governing equations and relevant constitutive models of the two-fluid models applied in the current research.

The mass conservation equation for the gas phase (q = g) and solid phase (q = s) is

$$\frac{\partial}{\partial t} (\varepsilon_g \rho_g) + \nabla \cdot (\varepsilon_g \rho_g \mathbf{v}_g) = \delta m_s$$

(1)

where $\delta m_s$ represents the mass of the gas produced by the reaction, $\mathbf{v}_g$ denotes the velocity of phase q, and $\varepsilon_q$ refers to the phase volume fraction.

Gas phase:

$$\frac{\partial}{\partial t} (\varepsilon_g \rho_g \mathbf{v}_g) + \nabla \cdot (\varepsilon_g \rho_g \mathbf{v}_g \mathbf{v}_g) = -\varepsilon_g \nabla P - \varepsilon_g \rho_g g + \beta_g (\varepsilon_g - \varepsilon_k) + \nabla \tau_g$$

(2)

Solid phase:

$$\frac{\partial}{\partial t} (\varepsilon_s \rho_s \mathbf{v}_s) + \nabla \cdot (\varepsilon_s \rho_s \mathbf{v}_s \mathbf{v}_s) = -\varepsilon_s \nabla P - \varepsilon_s \rho_s \gamma_s + \varepsilon_s \rho_s g + \beta_s (\varepsilon_s - \varepsilon_k) + \nabla \tau_s$$

(3)

where $\varepsilon_s = 1 - \varepsilon_g$.

The transport equation for granular temperature, $\theta$ (kinetic fluctuation energy of particles), is given as

$$3 \frac{\partial}{\partial t} (\varepsilon_s \rho_s \mathbf{v}_s \theta) + \nabla \cdot (\varepsilon_s \rho_s \mathbf{v}_s \mathbf{v}_s \theta) = (\nabla \rho_s T + \mathbf{v}_s : \nabla (k_s \nabla \theta) - \gamma_s + \phi_s + D_\theta)$$

(4)

The constitutive models used are as follows

Gas- and solid-phase stress tensors:

$$\tau_g = \varepsilon_g \mu \left[(\nabla \mathbf{v}_g + \nabla \mathbf{v}_g^T) - \frac{2}{3} \nabla \rho_g T \right]$$

(5)

$$\tau_s = \varepsilon_s \mu \left[(\nabla \mathbf{v}_s + \nabla \mathbf{v}_s^T) + \varepsilon_s \left(\frac{2}{3} \rho_s T \right) \nabla \mathbf{v}_s^T \right]$$

(6)

Radial distribution function:

$$g_0 = [1 - (\varepsilon_s / \varepsilon_s^{\text{max}})^{1/3}]^{-1}$$

(7)

Collisional energy dissipation:

$$\gamma_s = \frac{12(1 - \varepsilon_s^2) g_0}{d_s \sqrt{\pi}} \rho_s \varepsilon_s^2 \theta_s^{1/2}$$

(8)

Solid pressure:

$$p_s = \varepsilon_s \rho_s \theta + 2\rho_s (1 + \varepsilon_s) \varepsilon_s^2 \theta_s g_0$$

(9)

Solid shear viscosity:

$$\mu_{sk} = 4 \varepsilon_s^2 \rho_s d_s (1 + \varepsilon_s) \sqrt{\frac{\theta}{\pi}} + \frac{10 \rho_s d \sqrt{\pi} \theta}{96 (1 + \varepsilon_s) \varepsilon_s g_0}$$

$$\left[1 + \frac{4}{5} \varepsilon_s g_0 (1 + \varepsilon_s)^2 \right]^2$$

(10)

Frictional viscosity as given by Schaeffer et al. is
The diffusivity of granular temperature is given by

\[ k_{\theta\varepsilon} = \frac{150d_{\varepsilon}^2 \theta}{384(1 + c_{\varepsilon})g_0} \left[ 1 + \frac{6}{5} \varepsilon g_0(1 + c_{\varepsilon}) \right] \frac{\theta}{\pi^2} + 2\varepsilon d_{\varepsilon}^2 g_0(1 + c_{\varepsilon}) \frac{\theta}{\pi} \]  
(12)

Solid bulk viscosity is given as

\[ \varepsilon_{\varepsilon} = \frac{4}{3} \varepsilon d_{\varepsilon}^2 g_0(1 + c_{\varepsilon}) \frac{(\theta/\pi)^2}{150} \]  
(13)

The energy conservation equation is given as

\[ \frac{\partial}{\partial t} (\varepsilon d_{\varepsilon}^2 g_0(1 + c_{\varepsilon})) + \nabla \cdot (\varepsilon d_{\varepsilon}^2 g_0(1 + c_{\varepsilon}) \varepsilon) = -\frac{d P_q}{dt} + \nabla \cdot (\varepsilon d_{\varepsilon}^2 g_0(1 + c_{\varepsilon}) \varepsilon) + \sum_{q=1}^{n} (F_{pq} + m_{pq} v_{pq} - m_{pq} v_{pq} + S_q) \]  
(17)

In eq 17, \( h \) denotes the enthalpy (J/kg) of the \( q \) component. The experimental data involved in the study conducted by Ma et al.\textsuperscript{11–13} for a cylindrical spouted bed with a conical base, as shown in Figure 1b, are used to validate the numerical model of water vaporization and semidry flue gas desulfurization processes in a PPSB. The simulation deviation of desulfurization efficiency is 0.96%, indicating that the numerical model of desulfurization in the spouted bed is valid.
reasonable. The bed and grid structures for the computational domain are illustrated in Figure 2, along with the corresponding parameters selected for the present simulation in Table 1.

**Table 1. Size and Parameters of the Spouted Bed**

| description                  | computer run | description                  | computer run |
|------------------------------|--------------|------------------------------|--------------|
| diameter of the bed          | 53.5 mm      | minimum spouting velocity    | 0.56 m/s     |
| inclined angle               | 60°          | maximum solid volume fraction| 0.55         |
| bed height                   | 400 mm       | particle density             | 2700 kg/m³  |
| diameter of the spout gas    | 14.3 mm      | particle diameter            | 460 μm       |
| static bed depth             | 107 mm       | operating pressure           | 10 1325 Pa   |
| particle recovery coefficient| 0.9          | internal friction angle of particles | 28.7° |

2.2. Water Evaporation Model. 2.2.1. Model of Mass Transfer. In an environment with high-temperature gas, desulfurization slurry is wrapped on the surface of moving particles and evaporated, along with the application of the UDF method to realize water heat and mass transfer by considering evaporation in the simulation process. Based on the following hypothesis, a new model describing the water vaporization process is derived from a model of fuel droplet vaporization.

(a) As shown in Figure 3, the entire mass transfer in the water vaporization process is divided into two layers. Inner layer

![Figure 3. Simplified picture of the water evaporation model.](image)

I is a near-interface vaporization layer, and its thickness is extremely thin. Outer layer II is a diffusion layer that occupies most of the mass-transfer film.

(b) The gas temperature of inner layer I is uniform and equal to liquid film temperature because it is extremely thin and the specific heat of water is much greater than the specific heat of gas.

(c) The mass-transfer driving force of near-interface vaporization layer I is determined by the relative humidity gradient in the mass-transfer film. However, the mass-transfer driving force of diffusion layer II is dominated by the mass concentration gradient of the vaporization component.

The water evaporation rate of per unit area of desulfurization slurry is defined as

\[ m_w = \frac{D_g}{2R_d}Sh \rho_{total}B_M \]  

where \( D_g \) denotes the gas diffusion rate, \( R_d \) refers to the particle radius, and \( \rho_{total} \) represents the total density of the gas phase for the gas–liquid interface (kg/m³). \( Sh \) refers to the Sherwood dimensionless number, defined as

\[ Sh = (2.0 + 0.6Re^{1/2}Sc^{1/3})(1 + B_M)^{-0.7} \]

where \( Re \) represents the Reynolds number of droplets, \( Sc \) denotes the Schmidt dimensionless number, and \( B_M \) refers to the difference between the mass fraction of saturation vapor in the gas–liquid interface and that of water vapor in the gas phase subject value, which can be expressed as

\[ Re = \frac{d_P \rho_l \left| v_g - v_l \right|}{\mu_w} \]

\[ Sc = \frac{\mu_w}{\rho_l D} \]

\[ B_M = \frac{Y_{sat}(1 - \varphi)}{1 - Y_{sat}} \]

The expression of relative humidity, \( \varphi \), in the gas phase is as follows

\[ \varphi = \frac{P_{V2O}}{P_{sat}(MIN(T_w, T_g))} \]

where \( P_{sat}(MIN(T_w, T_g)) \) refers to a vapor-saturated vapor pressure function, whose value is calculated by the smaller values of liquid and gas temperature. The area of mass transfer between phases per unit volume is expressed as

\[ A_T = \alpha_f \alpha_s \frac{6e_{\text{pc}}}{d_{\text{pc}}} \]

The liquid film coating degree, \( \alpha_s \), is expressed as

\[ \alpha_s = \text{MIN} \left( \frac{\varepsilon_w d_{\text{pc}}}{12e_{\text{pc}} d_{\text{pf}}}, 1 \right) \]

where \( \varepsilon_w \) refers to the effect exerted by the volume fraction of the aqueous phase on the mass-transfer area, expressed as

\[ \alpha_w = \begin{cases} 1.0 & \varepsilon_w \leq 0.0001 \\ \frac{1}{100 000(\varepsilon_w - 0.0001) + 1} & \varepsilon_w > 0.0001 \end{cases} \]

where \( \varepsilon_w \) represents the volume fraction of the aqueous phase. The product of water vaporization rate, \( m_w \), and mass-transfer area, \( A_T \), serves as the source term \( \dot{m} \) of water vaporization mass transfer, defined as

\[ \dot{m} = m_w A_T \]

2.2.2. Model of Heat Transfer. The impact exerted by the latent heat of vaporization of water is considered. The gas-phase energy source term is defined as

\[ S_{\text{he}} = \dot{m}_{\text{vap}} h_{\text{vap}}(T_w, T_g) \]
Transfer between units of volume between two phases as follows:

The desulfurization process can be expressed in the following steps:

1. The SO\(_2\) in the high-temperature flue gas diffuses from the gas phase to the gas–liquid interface.
2. The SO\(_2\) diffuses from the gas–liquid interface into the liquid phase, beginning to be dissolved on the liquid membrane surface.
3. The SO\(_2\) dissolving in the water, generates H\(_2\)SO\(_3\), followed by dissociation.
4. The S ion in the liquid film diffuses to the center of the liquid phase.

The overall reaction equation is expressed as

\[ \text{SO}_2(\text{g}) + \text{Ca(OH)}_2(\text{s}) \rightarrow \text{CaSO}_3\cdot\frac{1}{2}\text{H}_2\text{O}(s) + \frac{1}{2}\text{H}_2\text{O}(l) \]

The definition of desulfurization efficiency is

\[ \eta_{\text{SO}_2} = \frac{C_{\text{in},\text{SO}_2} - C_{\text{out},\text{SO}_2}}{C_{\text{in},\text{SO}_2}} \]

In addition, assumptions for the momentum exchange between phases are listed as follows.

(a) The aqueous phase forms a liquid film that coats the particle surface, so that the drag force between water and gas phases can be neglected. The drag coefficient between gas–solid and liquid–solid phases is, respectively, determined by the Gidaspow model and symmetric model.

(b) The desulfurization phase and water phase constitute a wet desulfurization slurry, that is why the drag force between desulfurizer and gas phases is also neglected, and the drag coefficient between the desulfurizer and particle phases and water phase adopts a symmetric model.

### Table 2. Each Component Material and Physical Property Parameter of Inlet Gas

| chemical formula of components | H\(_2\)O | SO\(_2\) | O\(_2\) | N\(_2\) |
|-------------------------------|---------|---------|--------|--------|
| density (kg/m\(^3\))          | 0.5542  | 2.77    | 1.2999 | 1.138  |
| heat capacity at constant pressure [J/(kg·K)] | 2014 | 622.28  | 919.31 | 1040.67 |
| coefficient of thermal conductivity [W/(m·K)] | 0.0261 | 0.0104  | 0.0246 | 0.0242 |
| dynamic viscosity [kg/(m·s)] | 1.34 \times 10^{-3} | 1.2 \times 10^{-3} | 1.919 \times 10^{-5} | 1.663 \times 10^{-5} |
| molar mass (kg/kmol)          | 18.01534 | 64.0648 | 120.1442 | 28.0134 |
| standard enthalpy of formation (J/kmol) | 2.418379 \times 10^5 | 2.968612 \times 10^5 | 0 | 0 |
| reference temperature (K)     | 298     | 298     | 298    | 298    |

### Table 3. Each Physical Property Parameter of Other Components

| material                  | water phase | desulfurizer | product | particle |
|---------------------------|-------------|--------------|---------|----------|
| density (kg/m\(^3\))      | 998.2       | 2248         | 1595    | 2700     |
| heat capacity at constant pressure [J/(kg·K)] | 4182 | 1181.22     | 761.26  | 742.29   |
| coefficient of thermal conductivity [W/(m·K)] | 0.6  | 2.25         | 0.5     | 1.4      |
| dynamic viscosity [kg/(m·s)] | 0.001003 | 1.72 \times 10^{-3} | 1.72 \times 10^{-3} | 1.72 \times 10^{-5} |
| molar mass (kg/kmol)      | 18.01534    | 74.09268     | 120.1442 | 60.0843  |
| standard enthalpy of formation (J/kmol) | 2.858412 \times 10^4 | 9.8609 \times 10^6 | 1.71752 \times 10^4 | 9.1086 \times 10^4 |
| reference temperature (K) | 298         | 298          | 298     | 298      |

### Table 4. Boundary Conditions for Numerical Simulation

| initial and boundary conditions | parameter |
|-------------------------------|-----------|
| inlet of gas                  | turbulent velocity distribution; spouting inlet gas velocity \( U \), (m/s); turbulence kinetic intensity = 2%; hydraulic diameter = 0.0143 m; \( T = \) constant |
| inlet of desulfurization slurry | no particles enter for the solid phase; inlet gas temperature \( T = T_{\text{solid}} \), the mass fraction of each component of the inlet gas is H\(_2\)O = 0, SO\(_2\) = 0.00118, O\(_2\) = 0.23264 |
| outlet                        | velocity = 0.0260 m/s; the direction of velocity is a vertical boundary surface; \( T = 300 \) K (calcium hydroxide aqueous solution) |
| wall                          | no slip boundary condition at the lateral bed wall is assumed for the gas phase, the boundary condition of Johnson and Jackson\(^{34}\) is used for the solid phase, and standard wall functions for near-wall treatment and adiabatic boundary condition for the energy transport equation |
| symmetrical axis              | axisymmetric boundary |

The liquid energy source term is expressed as

\[ S_{\text{h,}\text{w}} = -n_{\text{vap}} h_{\text{vap}} (T_{\text{w}}, T_{\text{g}}) \]  \hspace{1cm} (29)

Heat transfer occurs among gas, liquid, and solid phases in the process of water vaporization. The heat-transfer model adopted in this paper is proposed by Ranz and Marshal\(^{35,36}\) with the heat transfer between units of volume between two phases as follows

\[ Q_{\text{p},\text{j}} = \frac{k \cdot N_u}{d_{\text{pc}}} A_{\text{T}} \cdot \Delta T \]  \hspace{1cm} (30)

where the Nusselt number, \( N_u \), is expressed as

\[ N_u = 2 + 0.6 R_e^{1/2} P_r^{1/3} \]  \hspace{1cm} (31)

2.3. Desulfurization Model. The reaction of the desulfurization process can be expressed in the following steps:

(a) The SO\(_2\) in the high-temperature flue gas diffuses from the gas phase to the gas–liquid interface.

(b) The SO\(_2\) diffuses from the gas–liquid interface into the liquid phase, beginning to be dissolved on the liquid membrane surface.

(c) The SO\(_2\) dissolving in the water, generates H\(_2\)SO\(_3\), followed by dissociation.

(d) The S ion in the liquid film diffuses to the center of the liquid phase.

The dissolved ionized process of desulfurizer Ca(OH\(_2\)).
33 The product phase after desulfurization is taken out of the bed by gas stream. Therefore, only the drag effect between the product phase and gas phase and particle phase is considered and the Wen and Yu model and the symmetric model are, respectively, used for calculation.

2.4. Simulation Method. The CFD code Fluent 15 is utilized to simulate the hydrodynamics and desulfurization process in PPSB. A second-order upwind discretization scheme is applied for momentum, turbulence kinetic energy, and turbulence dissipation rate equations, along with the application of the phase-coupled PC-SIMPLE algorithm for pressure-velocity coupling. Transient simulations are performed with a constant time step of $1 \times 10^{-5}$ s. The convergence criteria for the solution are that all variable residuals, such as velocity, are less than $1 \times 10^{-3}$; with the geometry, boundary condition, and grids of the two-dimensional spouted beds shown in Figure 2. A structured grid is used in the calculation domain. A grid independence test is performed for the two-dimensional computational model of spouted bed, finding that the grid number of 13 457 is sufficiently fine to ensure a grid-independent solution, as demonstrated in Figure 1a. Hence, the total number of grid cells is set as 13 475. Each component material and physical property parameters of gas are listed in Table 2, with physical property parameters of other components in Table 3. The model was computed by two Intel Xeon E5-2699v4 CPU @2.2 GHz and a Nvidia Quadro K620 2GB DDR3 graphics card.

The boundary conditions for numerical simulations are listed in Table 4. A comparative investigation on the velocity of the particle phase between numerical simulation results and experimental data, as reported by Wu et al., has been made. The maximum deviations of particle velocity and gas-phase volume fraction in the spouted bed are less than 26 and 6.9% between numerical simulation and experimental data, respectively. The deviation is considered to be acceptable, which can be attributed to the simulation of the PPSB.

3. RESULTS AND DISCUSSION

3.1. Particulate Flow Characteristics. The gas–solid flow pattern, velocity, and composition distribution were presented before discussion on the effects of the operating condition in PPSB. Contours of the solid volume fraction in the bed for different values of the superficial velocity at $t = 5$ s are present in Figure 4. As the superficial gas velocity increases, the height of the fountain gradually increases and the passage of gas in the spout zone becomes wider, that is, the spout diameter increases. Figure 5 shows the contours of particle velocity in the bed for different values of the superficial velocity at $t = 5$ s. As shown in

(c) Figures 5a, in the fountain area, the particles move downward, thus resulting in the axial velocity of the particles in this region is negative. However, in the annulus region, the solid volume fraction is large, and the gas and particles move in opposite directions, making the axial velocity of the particles in this region small. Figure 5b shows the radial velocity distribution of particles. The radial velocity of particles in the fountain zone and the annulus zone increases with the increase in superficial gas velocity. As the superficial gas velocity increases, the degree of turbulence of the gas phase and the particle phase increases, which effectively promotes the radial mixing between the gas and solid phases.

Axial distributions of the particle volume fraction for different values of the superficial velocity are compared in Figure 6. It can be seen that the particle volume fraction at nozzle is zero, increases with the height of bed, and finally decreases sharply to zero. This is because particles move upward under the drag provided by gas and accumulate continuously in the beginning. When particles rise to a certain height, the drag force is less than the gravity and particles start to decelerate upward. The particle
volume fraction reduces until the velocity is zero and then falls back from both sides to form a fountain zone, at which the particle volume fraction is minimized.

3.2. Effect of Operating Temperature on Desulfurization Efficiency. To further verify the reliability of the numerical model, the comparison in the temperature distribution at each phase of the PPSB when the gas inlet temperature, as demonstrated in Figure 7, is set as 520 K is made. As shown in Figure 7a, the temperature at the gas inlet is the highest, followed by the spout and the surrounding bed, and the lowest in the annulus and the fountain zone. This reveals that heat transfer occurs among gas, water, and particle phases when the high-temperature flue gas enters the spouted bed, with the increase in bed height, thus resulting in the decrease in gas temperature in the spout region. However, due to the high gas velocity and short heat transfer time in this region, the temperature of the gas decreases insignificantly. However, in the annulus region of the spouted bed, the air flow is large, the contact time among gas, particles, and water is enough, helping explain that the heat transfer process is sufficient, and the temperature of the gas decreases significantly. In addition, the temperature distributions of water and particle phases in Figure 7b,c are similar to those in the gas phase (Figure 7a), which is because the momentum and heat transfer between the multiphase in PPSB are driven by the gas phase. The results show the best mass and heat transfer effect of each phase in the outer annulus and the near spout region, both of which are also the main areas where water vaporization occurs. Also, the simulation results of Figure 7 show that the numerical simulation model adopted in this paper is reliable.

In the process of PPSB, the removal reaction of SO₂ is an exothermic reaction, which is the core link of the flue gas desulfurization process. Temperature, as a nonnegligible factor, is of important research value. Excessive temperature will change the solubility of the gas and absorbent, thus affecting the SO₂ removal effect. The low temperature makes the flue gas difficult to diffuse, incapable of meeting the standard of flue gas emission. Therefore, it is particularly important for the industrial desulfurization process to study the appropriate temperature range of inlet gas so that spouted bed can not only have higher desulfurization efficiency but also meet the requirements of flue gas emission. Due to the sensitivity of the operating temperature to desulfurization efficiency in PPSB, the desulfurization process and desulfurization efficiency in the spouted bed at different operating temperatures (T = 480, 500, 520, 540, and 560 K) were analyzed by the CFD model when the superficial gas velocity was kept constant as \( U = 1.2 \, U_{max} \). Figure 8 shows the vaporization rate in the spouted bed for different temperatures of inlet flue gas, finding that the main areas of water vaporization are the spout zone and outer annulus. The rate of water vaporization increases with the increase in inlet gas temperature. Because the water content of the slurry is constant, the water of slurry gets more heat from the gas phase, along with the rise of its temperature when the inlet temperature increases, accelerating the rate of water vaporization. Therefore, the rate of water vaporization will increase with the increase in inlet temperature, especially in the spout zone.

Figure 9 shows the desulfurization product (CaSO₃) rate in the spouted bed for different values of the temperature of inlet gas. It can be seen from Figure 9 that the rate of desulfurization products decreases with the increase in inlet gas temperature, indicating that, on the one hand, the increase in temperature of imported flue gas is accompanied by that of the rate of water vaporization, that is, more water vaporizes. Water is an important medium in the desulfurization reaction, and the solubility of the absorbent and sulfur dioxide in water decreases when the amount of water decreases, thus making the rate of desulfurization reaction lower. On the other hand, the increase in temperature decreases the solubility of gas, thereby increasing the driving force of gas-film mass transfer. Hence, more gas is dissolved at the gas–liquid interface through the gas film, further increasing the possibility of contacting with the absorbent and the reaction area, along with the increase in reaction rate of desulfurization. In consideration of the two above factors, it can be concluded that the desulfurization reaction rate decreases with the increase in temperature, that is, the desulfurization process is mainly affected by the dissolution process of the absorbent and sulfur dioxide.

Figure 10 further displays the volume fraction distribution of the desulfurization reaction product (CaSO₃) in the spouted bed for different temperatures of the inlet flue gas, clearly showing that the volume fraction of the desulfurization reaction product decreases with the increase in temperature.
products decreases with the increase in inlet flue gas temperature. The volume fraction of the desulfurization reaction products is the largest when the inlet flue gas temperature is 480 K, which is $2.91 \times 10^{-5}$. The volume fraction of desulfurization reaction products is correspondingly related to the desulfurization process. The absorption of SO$_2$, an exothermic reaction, is directly related to temperature. Too high temperatures will inhibit the absorption of SO$_2$ and decompose the desulfurization products. Therefore, the increase in temperature will not be conducive to the reaction in the direction of product formation.

Figure 11 shows the comparison in desulfurization efficiency in the spouted bed for different temperatures of inlet flue gas, implying that the highest desulfurization efficiency, compared with other working conditions, is 84.23% when the inlet flue gas temperature is 480 K. When the temperature of inlet flue gas rises, the desulfurization efficiency decreases with a certain amount of imported slurry and water content, but the temperature should not be too low. Solid particles are easy to bond and agglomerate at a low temperature, along with the fouling on the wet wall caused by the incomplete evaporation of water, which is not conducive to the emission of gas, thus bringing trouble for the later treatment of flue gas and cleaning of equipment. Also, excessive temperature cannot achieve the expected desulfurization effect, along with the corresponding increase in the cost. Therefore, to obtain a better desulfurization rate in PPSB, the temperature of the inlet flue gas should be controlled as low as possible above the minimum limit.

3.3. Effect of Superficial Velocity on Desulfurization Efficiency. The vaporization rate in the bed for different values of the superficial velocity at $t = 5$ s when the operating temperature of inlet gas is kept constant as $T = 520$ K is compared, as demonstrated in Figure 12. Interestingly, the vaporization rate of water decreases gradually with the increase in superficial gas velocity, playing a key role in the desulfurization process. The residence time of the gas in the spouted bed becomes shorter with the increase in superficial gas velocity, causing the gas to leave the bed without sufficient heat transfer with water, and the water does not reach the temperature required for vaporization, thus decreasing the vaporization rate of water. The vaporization rate along the radial direction in the bed for different values of the superficial velocity at different bed heights is compared in Figure 13, demonstrating that under different superficial gas velocities the water vaporization rate decreases slowly with the increase in radial distance of PPSB. To be more specific, when the bed height is low ($z = 0.03$ m), the superficial gas velocity $U = 1.0$ and $1.1 U_{ms}$ along with the highest water vaporization rate in the spouted bed. This indicates that the contact time of gas, particle, and liquid water in the spouted bed is longer when the superficial gas velocity is lower and the bed height is close to the nozzle, where the gas temperature is high, which can be conducive to the vaporization process of water in PPSB. To be more specific, the water vaporization rate increases first and then decreases with the increase in superficial gas velocity when the superficial gas velocity is greater than or equals to $1.2 U_{ms}$. With the increase in bed height ($z = 0.06, 0.09$ m), which is far from the gas inlet, and when the gas-phase temperature is lower, the impact exerted by the superficial gas velocity on the water vaporization rate in the spouted bed decreases gradually, with the values of water vaporization rate in five working conditions being small and not very different from each other.

Figure 14 shows the comparison of the desulfurization product (CaSO$_3$) rate along the radial direction in the bed for different values of the superficial velocity. It can be seen from Figure 14 that the behavior of the desulfurization product rate along the spouted bed radial position is similar under different superficial gas velocities, with the appearance of the peak desulfurization efficiency in the annulus. However, the superficial gas velocity increases with the increase in turbulence energy among three phases of gas, liquid, and solid and mass-transfer coefficient, which is beneficial for the process of desulfurization in PPSB. However, at the same time, the reaction time is shorter and the absorption of SO$_2$ is not complete, thus reducing the desulfurization rate. The residence time of flue gas in the spouted bed serves as the main factor affecting the flue gas desulfurization process and takes up a dominant position. A comprehensive analysis of the above two factors demonstrates that the desulfurization product rate is negatively correlated with the change of superficial gas velocity.

Figure 15 compares the volume fraction distribution of the desulfurization reaction product (CaSO$_3$) in the bed for different values of the superficial velocity, indicating that the
Figure 13. Comparison of the vaporization rate along the radial direction in the bed for different values of the superficial velocity [unit: kg/(m$^3$ s)]. (a) $z = 0.03$ m, (b) $z = 0.06$ m, and (c) $z = 0.09$ m.

Figure 14. Comparison of the desulfurization product (CaSO$_3$) rate along the radial direction in the bed for different values of the superficial velocity [unit: kg/(m$^3$ s)]. (a) $z = 0.03$ m, (b) $z = 0.06$ m, and (c) $z = 0.09$ m.
The variation of the desulfurization efficiency in PPSB and obtain considerable processing capacity. Therefore, the optimal superficial gas velocity equals 1.2 \( U_{\text{ms}} \) and the desulfurization efficiency and gas handling capacity reach the best result, which can contribute to the optimum design of PPSB.

4. CONCLUSIONS

(1) The mass and heat transfer effect of each phase in the outer annulus and the near spout region is the highest, both of which are also the main areas where water vaporization occurs. The increase in inlet gas temperature is followed by that of the water vaporization rate, while that of desulfurization products decreases with the increase in inlet gas temperature. Even more, the volume fraction of desulfurization reaction products decreases with the increase in inlet flue gas temperature.

(2) Compared with other working conditions, the highest desulfurization efficiency reaches 84.23\% when the inlet flue gas temperature is 480 K. Besides, the desulfurization efficiency decreases with a certain amount of imported slurry and water content when the temperature of inlet flue gas rises.

(3) The increase in superficial gas velocity is accompanied by a gradual decrease in vaporization rate of water. When the bed height is low (\( z = 0.03 \) m), the superficial gas velocity \( U = 1.0 \) and \( 1.1 \) \( U_{\text{ms}} \) and the water vaporization rate in the spouted bed is the highest. The change rule of desulfurization product rate with the radial distance is the same under different superficial gas velocities, with the appearance of the peak desulfurization efficiency in the annulus.

(4) The optimal selection point for the desulfurization process in PPSB is available. The desulfurization rate and the processing capacity are higher when the superficial gas velocity equals \( 1.2 \) \( U_{\text{ms}} \) and the desulfurization efficiency and gas handling capacity reach the best result, which can contribute to the optimum design of PPSB.

### AUTHOR INFORMATION

**Corresponding Author**

Feng Wu – School of Chemical Engineering, Northwest University, Xi’an 710069, China; orcid.org/0000-0002-8943-4926; Phone: +86-15309202861; Email: wufeng@nwu.edu.cn

---

**Figure 15.** Volume fraction distribution of the desulfurization reaction product (CaSO\(_3\)) in the bed for different values of the superficial velocity.

**Figure 16.** Comparison of the desulfurization efficiency and outlet gas flow rate in PPSB for different values of the superficial velocity \((T = 520 \text{ K})\). (a) Desulfurization efficiency vs superficial velocity and (b) desulfurization efficiency and outlet gas flow rate vs superficial velocity.
Authors
Jinhao Bai — School of Chemical Engineering, Northwest University, Xi’an 710069, China
Kai Yue — School of Chemical Engineering, Northwest University, Xi’an 710069, China
Ming Gong — School of Chemical Engineering, Northwest University, Xi’an 710069, China
Xiaoxun Ma — School of Chemical Engineering and Technology, Xi’an Jiaotong University, Xi’an 710049, China
Wenjing Zhou — School of Chemical Engineering and Technology, Xi’an Jiaotong University, Xi’an 710049, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.9b03361

Notes
The authors declare no competing financial interest.

ACKNOWLEDGMENTS
This work is financially supported by the National Natural Science Foundation of China (Grant no. 21878245) and the Natural Science Foundation of Shaanxi Province (Grant no. 2019JM-039).

NOMENCLATURE
\( C \) [kg/m\(^3\)] concentration  
\( d \) [mm] particle diameter  
\( d_p \) [mm] droplet diameter  
\( D \) [m/s\(^2\)] diffusion coefficient  
\( B_M \) — spalding mass number  
\( g_0 \) — radial distribution function  
\( h \) [J/kg] enthalpy  
\( H \) [mm] vessel height  
\( m \) [kg/(m\(^3\)*s)] mass transfer  
\( m_a \) [kg/(m\(^2\)*s)] water evaporation rate per unit area  
\( t \) [s] time  
\( T \) [K] thermodynamic temperature  
\( R_e \) — Reynolds dimensionless number  
\( R_{e_p} \) Reynolds number of particles  
\( S \) — generalized source term  
\( Sc \) — Schmidt dimensionless number  
\( Sh \) — Sherwood dimensionless number  
\( U \) [m/s] superficial gas velocity  
\( U_{ms} \) [m/s] minimum spouting velocity  
\( v \) [m/s] velocity  
\( x, y \) [m] Cartesian coordinates  
\( Y_{sat} \) — gas–liquid interface vapor mass fraction

GREEK SYMBOLS
\( \beta_p \) [kg/(m\(^3\)*s)] solid–gas momentum exchange coefficient  
\( \varepsilon \) [—] volume fraction  
\( \theta \) [m\(^2\)/s\(^3\)] granular temperature  
\( \rho \) [kg/m\(^3\)] density  
\( \rho_w \) [kg/m\(^3\)] density of water  
\( \mu \) [kg/(m\(^3\))*s] shear viscosity  
\( \mu_w \) [kg/(m\(^3\)*s)] shear viscosity of water  
\( \xi \) [kg-H\(_2\)O/kg] water content of slurry  
\( \eta \) [%] desulfurization efficiency

SUBSCRIPTS
\( g \) [—] gas  
\( q \) [—] phase type (solid or gas)  
\( s \) [—] solids

REFERENCES
1. García-Labiano, F.; Rufas, A.; de Diego, L. F.; de las Obras-Loscertales, M.; Gayán, P.; Abad, A.; Adánez, J. Calcium-based sorbents behaviour during sulphaion at oxy-fuel fluidised bed combustion conditions. *Fuel* 2011, 90, 3100–3108.
2. Jia, L.; Tan, Y.; Wang, C.; Anthony, E. J. Experimental Study of Oxy-Fuel Combustion and Sulfur Capture in a Mini-CFBC. *Energy Fuels* 2007, 21, 3160–3164.
3. Mohanty, C. R.; Adapala, S.; Meikap, B. C. Removal of hazardous gaseous pollutants from industrial flue gases by a novel multi-stage fluidized bed desulfurizer. *J. Hazard. Mater.* 2009, 165, 427–434.
4. Zhang, Q.; Gui, K. A novel semidry flue gas desulfurization process with the magnetically fluidized bed reactor. *J. Hazard. Mater.* 2009, 168, 1341–1345.
5. Tao, M.; Jin, B.; Zhong, W.; Yang, Y.; Xiao, R. Modeling and experimental study on multi-level humidifying of the underfeed circulating spouted bed for flue gas desulfurization. *Powder Technol.* 2010, 198, 93–100.
6. Tao, M.; Jin, B.; Zhong, W.; Yang, Y.; Xiao, R. Numerical and experimental study on flue gas desulfurization in the underfeed circulating spouted bed. *Chem. Eng. J.* 2010, 159, 149–158.
7. Shun, D.; Bae, D. H.; Jang, I. K.; Park, K. H.; Park, S. K. Kinetic Study of Sulfur Dioxide Elimination by Limestone through the Lab Scale Circulating Fluidized Bed Combustor. *Adv. Mater. Phys. Chem.* 2012, 02, 189–192.
8. Wall, T.; Liu, Y.; Spero, C.; Elliott, L.; Khare, S.; Rathnam, R.; et al. An overview on oxyfuel coal combustion—state of the art research and technology development. *Chem. Eng. Res. Des.* 2009, 87, 1003–1016.
9. Lupion, M.; Diogo, R.; Loubeau, L.; Navarrete, B. CIUDEN CCS project: Status of the CO\(_2\) capture technology development plant in power generation. *Energy Procedia* 2011, 4, 5639–5646.
10. Qimin, G.; Noriaki, I.; Kato, K. Process development of effective semidry flue gas desulfurization by a powder-particle spouted bed. *Kagaku Kogaku Ronbunshu* 1996, 22, 1400–1407.
11. Xu, G.; Kaneko, T.; Guo, Q.; Xu, G.; et al. Removal of SO\(_2\) from flue gas using a new semidry flue gas desulfurization process with a powder-particle spouted bed. *Can. J. Chem. Eng.* 1999, 77, 356–362.
12. Wang, N.; Zhang, X.; Kaneko, T.; Tashimo, T.; Yoshida, T.; Kato, K. Use of limestone for SO\(_2\) removal from flue gas in the semidry FGD process with a powder-particle spouted bed. *Chem. Eng. Sci.* 2000, 55, 4643–4652.
13. Ma, X.; Kaneko, T.; Xu, G.; Kato, K. Influence of gas components on removal of SO\(_2\) from flue gas in the semidry FGD process with a powder-particle spouted bed. *Fuel* 2001, 80, 673–680.
14. Xu, G.; Guo, Q.; Kaneko, T.; Kato, K. A new semidry desulfurization process using a powder-particle spouted bed. *Adv. Environ. Res.* 2000, 4, 9–18.
15. Wu, S.; Uddin, A.; Su, C.; Nagamine, S.; Sasaoka, E. Effect of the pore-size distribution of lime on the reactivity for the removal of SO\(_2\) in the presence of high-concentration CO\(_2\) at high temperature. *Ind. Eng. Chem. Res.* 2002, 41, 5455–5458.
16. Wang, N.; Zhang, X. Effect of humidification water on semi-dry flue gas desulfurization. *Procedia Environ. Sci.* 2011, 11, 1023–1028.
17. Rahimi, A.; Hatamipour, M. S.; Gholami, M.; Haghnehadhar, M. R. Non-isothermal modeling of the flue gas desulfurization process using a semi-dry spouted bed reactor. *Chem. Eng. Res. Des.* 2011, 89, 777–784.
18. Fakhari, M. A.; Rahimi, A.; Hatamipour, M. S.; Fozooni, A. Non-isothermal modeling of simultaneous CO\(_2\) and SO\(_2\) removal in a semi-dry spouted bed reactor. *Process Saf. Environ. Prot.* 2015, 98, 342–353.
19. Haghnehadhar, M. R.; Hatamipour, M. S.; Rahimi, A. Removal of carbon dioxide in an experimental powder-particle spouted bed reactor. *Sep. Purif. Technol.* 2010, 72, 288–293.
20. Mathur, K. B.; Lim, C. J. Vapor phase chemical reaction in spouted beds: a theoretical model. *Chem. Eng. Sci.* 1974, 29, 789–797.
21. Ghalavand, Y.; Rahimi, A.; Hatamipour, M. S. Experimental study and mathematical modeling of green pea drying in a spouted bed. *Drying Technol.* 2012, 30, 128–137.
(22) Haghnegahdar, M. R.; Hatamipour, M. S.; Rahimi, A. Mathematical modeling of CO₂ capture in a semi-dry spouted bed reactor. *Sep. Purif. Technol.* 2011, 80, 509–518.

(23) Niksiar, A.; Sohrabi, M.; Rahimi, A. Comparative evaluation of existing correlation to predict spouted bed hydrodynamics. *Drying Technol.* 2013, 31, 975–989.

(24) Silva, D. O.; Tamiozzo, L. M.; Duarte, C. R.; Murata, V. V.; Barrozo, M. A. S. Modeling of seed coating in a spouted bed. *Drying Technol.* 2011, 29, 286–294.

(25) Fakhari, M. A.; Rahimi, A.; Hatamipour, M. S.; Fozooni, A. Experimental study of simultaneous removal of CO₂ and SO₂ in a spouted bed reactor. *Can. J. Chem. Eng.* 2017, 95, 1150–1155.

(26) Niksiar, A.; Nasernejad, B. Modeling of gasification reaction to produce activated carbon from pistachio shells in a spouted bed. *Biomass Bioenergy* 2018, 119, 97–108.

(27) Tseng, C. C.; Li, C. J. Eulerian-Eulerian numerical simulation for a flue gas desulfurization tower with perforated sieve trays. *Int. J. Heat Mass Transfer* 2018, 116, 329–345.

(28) Lun, C. K.; Savage, S. B.; Jeffrey, D. J.; Chepurniy, N. Kinetic theories for granular flow: inelastic particles in Couette flow and slightly inelastic particles in general flow field. *J. Fluid Mech.* 1984, 140, 223–256.

(29) Schaeffer, D. G. Instability in the evolution equations describing incompressible granular flow. *J. Differ. Equations* 1987, 66, 19–50.

(30) Gidaspow, D.; Bezburaah, R.; Ding, J. In *Hydrodynamics of Circulating Fluidized Beds, Kinetic Theory Approach*, Fluidization VII, Proceedings of the Seventh Engineering Foundation Conference on Fluidization, 1992; pp 75–82.

(31) Sazhin, S. S.; Elwardany, A. E.; Krutitskii, P. A.; Deprédurand, V.; Castanet, G.; Lemoine, F.; Sazhina, E. M.; Heikal, M. R. Multi-component droplet heating and evaporation: Numerical simulation versus experimental data. *Int. J. Therm. Sci.* 2011, 50, 1164–1180.

(32) Warych, J.; Szymanowski, M. Model of the Wet Limestone Flue Gas Desulfurization Process for Cost Optimization. *Ind. Eng. Chem. Res.* 2001, 40, 2597–2605.

(33) Wu, F.; Zhang, X.; Zhou, W.; Ma, X. Numerical simulation and optimization of hydrodynamics in a novel integral multi-jet spouted-fluidized bed. *Powder Technol.* 2018, 336, 112–121.

(34) Johnson, P. C.; Jackson, R. Frictional-collisional constitutive relations for granular materials with application to plane shearing. *J. Fluid Mech.* 1987, 176, 67–93.

(35) Ranz, W. E.; Marshall, W. R., Jr. Evaporation from Drops, Part I. *Chem. Eng. Prog.* 1952, 48, 141–146.

(36) Ranz, W. E.; Marshall, W. R., Jr. Evaporation from Drops, Part II. *Chem. Eng. Prog.* 1952, 48, 173–180.