



# Numerical Simulation of the Operating Conditions for the Reduction of Iron Ore Powder in a Fluidized Bed Based on the CPFD Method

Zi-wei Wan, Jin-yu Huang, Guo-min Zhu and Qi-yan Xu \*

School of Metallurgical Engineering, Anhui University of Technology, Ma'anshan 211156, China

\* Correspondence: qiyan.xu@163.com

**Abstract:** In this work, the computational particle fluid dynamics (CPFD) method is used to simulate the high-pressure visual fluidized bed experimental equipment independently designed and developed by the experimentation of the fluidized reduction process of iron ore powder. A numerical model for reducing iron ore fines in a three-dimensional fluidized bed is established, and the model is verified by combining numerical simulation and experimental testing. Moreover, the influences of different reducing factors on the reduction effect in the process of the fluidized reduction of iron ore fines are simulated in detail. Via the CPFD simulation of the fluidized reduction of iron ore fines, the optimal reduction pressure is found to be 0.2 MPa, and the optimal reducing gas is found to be H<sub>2</sub>. Moreover, the optimal gas velocity is 0.6 m/s, and the optimal reduction temperature is 923 K. This conclusion is consistent with the experimental measurements, so the simulation results can be used to verify the reliability of the optimal operating conditions.

Keywords: fluidized bed; CPFD; simulation; iron ore fines; metallization rate

# 1. Introduction

The fluidized direct reduction process has the advantages of a large gas-solid contact area, a uniform temperature and concentration, good heat and mass transfer conditions, and high operating efficiency [1–3]. However, due to the complex multiphase flow system, particles will be entrained by the airflow to cause material loss, and the particles will cause wear and tear to the equipment [4–6]; consequently, the scale-up of industrial-scale fluidized beds faces huge challenges. Experimental research and numerical simulation are the main ways to study gas-solid two-phase flow, and experimental measurement data are the basis of theoretical analysis. At present, relevant scholars have carried out large amounts of experimental measurements and research on the internal flow mechanism of the fluidized bed, such as the minimum fluidization velocity, bed height, and local particle aggregation [7–9]. However, due to the limitation of the experimental conditions and the complexity and uncertainty of gas-solid two-phase flow, it is difficult to obtain specific information at the micro-level, such as the particle motion state and gas distribution [10-12], and experimental measurement has the disadvantages of a large amount of engineering, a high cost, a long cycle, and limited measurement methods. With the continuous development of computer technology, numerical simulation has become an important means by which to study dense gas-solid two-phase flow, and plays an increasingly important role in supplementary experimental research [13–17]. Numerical simulation can be used to accurately calculate, predict, and study gas-solid two-phase flow, and to more intuitively obtain the process and details of the gas-solid flow on the particle and grid scales, which are difficult to measure in an experiment. Moreover, this method is not limited by the measurement means, and can yield rich micro-information about the core factors [18–20]. Furthermore, numerical simulation can be used to directly calculate equipment of any scale, and can provide prediction results for the development of new reaction equipment to aid in the



Citation: Wan, Z.-w.; Huang, J.-y.; Zhu, G.-m.; Xu, Q.-y. Numerical Simulation of the Operating Conditions for the Reduction of Iron Ore Powder in a Fluidized Bed Based on the CPFD Method. *Processes* **2022**, *10*, 1870. https://doi.org/10.3390/ pr10091870

Academic Editor: Ireneusz Zbicinski

Received: 18 August 2022 Accepted: 8 September 2022 Published: 16 September 2022

**Publisher's Note:** MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



**Copyright:** © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). optimal design of internals. Liang et al. [21] used the computational particle fluid dynamics (CPFD) model to simulate a two-dimensional fluidized bed, and explored the influences of various parameters in the simulation on the calculation results. The results showed that when meshing, refinement near the wall can significantly improve the accuracy of the results, and the solid-phase velocity in the tube can be better predicted. Lim et al. [22] used the CPFD method to study the influence of particle interaction, determined the flow characteristics of the fluidized bed according to the collision model and the change of the normal stress parameters of the particles, and verified that CPFD simulation is more effective and stable than previous computational fluid dynamics (CFD) in fluidized bed analysis. Jia et al. [12] established a three-dimensional numerical simulation of circulating fluidized bed combustion to simulate the pyrolysis and combustion of coke and volatile substances from gas–solid flow, heat and mass transfer, and chemical reaction. This method allows for the strong prediction of the combustion characteristics of a circulating fluidized bed.

At present, there is a vacancy in the simulation of the influences of the mass transfer, heat transfer, and chemical reaction processes in the fluidized bed on the gas-solid twophase movement at high temperatures [23–25]. Because there are no unified and reliable chemical reaction kinetic parameters that can accurately simulate changes in gas composition, the complex physical and chemical processes and reaction kinetic models in the fluidized bed cause significant differences between the gas composition determined by the simulation and experimental results [26–28]. However, in theory, it is difficult to verify the rationality of the parameters of the numerical model and the reliability of the prediction results [29–31]; to a large extent, they must be verified and determined via experimental data. Only a chemical reaction kinetic model with a wider application scope and higher reliability can more accurately predict the changes of the gas composition [32–34]. Therefore, in view of these problems, this study takes numerical simulation and experimental measurement as the main content, which is then supplemented by theoretical analysis, to carry out numerical simulation research on the state of reduced iron ore powder in a pressurized fluidized bed. Coupled with the gas-solid flow and reaction dynamics characteristics, a numerical model of reduced iron powder in a pressurized circulating fluidized bed is established to reveal the flow and reaction characteristics of the fluidized reduction of iron powder, and to explore the influences of the operating parameters on the gasification results. Barracuda software is used to calculate and study the influences of the temperature field, chemical reaction, reduction pressure, and linear gas velocity on the reduction effect in the fluidized bed reactor. By comparing the metallization rate measured in the experiment with the simulated metallization rate, the influences of different factors on the reduction of ore powder in the experiment are verified. The optimal operation conditions of the simulated fluidized reduction of iron ore powder are obtained via comprehensive analysis, which provides a theoretical basis for understanding the flow law of the fluidized bed, optimizing the process and operation, and broadening its industrial application range.

#### 2. Experimental Steps and Protocols

The reduced iron ore powder used in this experiment was Newman ore from Australia, the shielding gas was  $N_2$ , and the reducing gases were  $H_2$ , CO, and a mixture of  $H_2$  and CO. The main device was a pressurized visible fluidized bed (Figure 1) with double stainless-steel tubes as the reactor, and the inner tube was a fluidized bed. First,  $N_2$  was introduced into the fluidized bed to discharge all the air and raise the pressure to 0.5–0.6 MPa to check the air tightness of the device. After heating the fluidized bed to the set temperature, the  $N_2$  valve was closed, and the reducing gas was introduced. The gas was regulated by a flowmeter and gas mass flow controller to allow the gas to enter the bed to reach the required linear speed and control the inlet flow of the gas. The experiment was initiated after the pressure stabilized to the set pressure value. A quantitative amount of fine mineral powder particles (500 g) was placed into the fluidized bed and reacted with the reducing gas for the set time, and then the experiment was stopped. After cooling to room

temperature, the mineral powder in the fluidized bed was removed, and the samples were analyzed by the potassium dichromate volumetric method and the ferric chloride titration method to determine the contents of metallic iron ( $M_{Fe}$ ) and total iron ( $T_{Fe}$ ) and calculate the metallization rate  $\eta$ . The higher the metallization rate, the better the quality of the reduced mineral powder. To reduce the experimental error, the measurement was repeated twice, and the average value was taken as the final experimental result. The experimental scheme is shown in Table 1.



**Figure 1.** Experimental setup: (**a**) fluidized bed reactor; (**b**) flow chart. 1. Gas mixing and preheating chamber; 2. Gas mixing hole; 3. Fluidized bed; 4. K-type thermal couple; 5. Pressure sensor; 6. Gravity filter; 7. Feeding and sampling port; 8. Pressure seal cap; 9. Temperature change recorder; 10. Gas analyzer; 11.  $H_2$  gas analysis recorder; 12.  $CO_2$  gas analysis recorder; 13. CO gas analysis recorder; 14. Pressure change analysis recorder; 15. Computer; 16. Gas dryer; 17. Gasholder; 18. Pressure display; 19.  $N_2$  gas cylinders; 20.  $CO/CO_2$  gas cylinders; 21.  $H_2$  gas cylinders; 22. Gas mass flowmeter; 23. Booster pump; 24. Gas valve.

No.	<b>Reduction Temperature/K</b>	Type of Reducing Gas	<b>Reducing Pressure/MPa</b>	Linear Velocity/m/s
1	923	H <sub>2</sub>	0.2	0.6
2	1023	H <sub>2</sub>	0.2	0.6
3	1123	H <sub>2</sub>	0.2	0.6
4	923	H <sub>2</sub>	0.1	0.6
5	1023	H <sub>2</sub>	0.1	0.6
6	1123	H <sub>2</sub>	0.1	0.6
7	1023	H <sub>2</sub>	0.1	0.6
8	1023	CO	0.1	0.6
9	1023	$H_2 + CO$	0.1	0.6
10	1023	H <sub>2</sub>	0.2	0.6
11	1023	CO	0.2	0.6
12	1023	$H_2 + CO$	0.2	0.6
13	1023	$H_2 + CO$	0.1	0.6
14	1023	$H_2 + CO$	0.2	0.6
15	1023	$H_2 + CO$	0.4	0.6
16	923	$H_2 + CO$	0.1	0.6
17	923	$H_2 + CO$	0.2	0.6
18	923	$H_2 + CO$	0.4	0.6
19	1023	$H_2 + CO$	0.1	0.4
20	1023	$H_2 + CO$	0.1	0.6
21	1023	$H_2 + CO$	0.1	0.8
22	1023	H <sub>2</sub>	0.2	0.4
23	1023	H <sub>2</sub>	0.2	0.6
24	1023	$H_2$	0.2	0.8

The metallization rate is calculated as follows:

$$\eta = M_{F_e} / T_{Fe} \tag{1}$$

where  $\eta$  is the metallization rate,  $M_{Fe}$  is metallic iron (g), and  $T_{Fe}$  is total iron (g).

#### 3. Mathematical Models

## 3.1. Governing Equations

In Barracuda Virtual Reactor 17.4.0 software, large eddy simulation (LES) was used for the turbulence model of the gas field, the MP-PIC method was used for particles in the solid phase, the normal stress of particle collision and the gas-solid heterogeneous reaction were calculated based on the Euler grid, and the properties of discrete particles were interpolated to the Euler grid. The solution was then interpolated back to the particles, and this process was realized by the interpolation operator. In order to make the grid size of the fluid phase larger than that of the particle phase, the total number of grids was finally determined to be 69,584 by grid-independence analysis when dividing the fluidized bed grid model, and the grid size was  $3.1579 \times 3.1707 \times 3.1098$ , which had the least effect on the pressure and temperature variations in the simulation. The drag force between the gas and solid was calculated by the Wen-Yu model, and the "calculated particles" with the same properties were packaged. Chemical reactions were combined with the complex hydrodynamics calculations of gas–solid fluidized beds. The setting of an "equivalent particle size" was adopted for the method based on CPFD, and small particles clustered to form large particles, which were replaced by equivalent particles when solving the drag force. The calculation method and fluidization phenomenon of large particles were revealed, and the thermal conductivity effect brought by agglomeration was simultaneously simulated. The fluidized bed model in the simulation was established and simplified according to the reaction principle presented in Figure 1. The main part was a cylinder with a height of 0.78 m and a diameter of 0.30 m. The system grid is presented in Figure 2. The main air intake was located at the bottom of the entire system. The origin was set at the center of the bottom of the riser, and the z-axis was opposite to the direction of gravity. An overview of the main governing equations is provided in Table 2.



Figure 2. Grids: (a) Overall Grid; (b) Grid 1; (c) Grid 2.

 $\begin{array}{c|c} \mbox{Equation} & \mbox{Equation Expression} \\ \hline Continuity equation & & \nabla(\rho_g a_g u_g) + \frac{\partial}{\partial t}(\rho_g a_g) = \delta \dot{m}_{s'} - \int \int \int \frac{dm_s}{dt} dT_s du_s dm_s = \delta \dot{m} \\ -a \nabla p + \nabla \tau_g + \rho_s a_g u_g g + F = \nabla(\rho_s a_g u_g u_g) + \frac{\partial}{\partial t}(\rho_g a_g u_g) \\ (\rho_g a_g u_g) + \nabla \tau_g + \rho_s a_g u_g g + F = \nabla(\rho_s a_g u_g u_g) + \frac{\partial}{\partial t}(\rho_g a_g u_g) \\ \delta \dot{m}_{g,i} + \delta \dot{m}_{s,i} + \nabla(\rho_g a_g D_g Y_{g,i}) = \nabla(\rho_s a_g Y_{g,i} u_g) + \frac{\partial}{\partial t}(\rho_g a_g Y_{g,i}) \\ \partial \partial t(a_g \rho_g h_g) + \nabla(a_g \rho_g h_g u_g) = a_g \left(\frac{\partial p}{\partial t} + u_g \nabla p\right) + \phi - \nabla(a_g q) + \dot{Q} + S_h + \dot{q}_D \\ Particle equation of motion \\ Particle collision model \\ Particle energy transfer model \\ Volume fraction of particles in unit grid \\ Local fluid wall heat transfer coefficient \\ Diluted phase heat transfer coefficient \\ Diluted phase heat transfer coefficient \\ Dirag coefficient \\ Drag coefficie$ 

#### Table 2. Governing equations [35–38].

#### 3.2. Chemical Reactions

All elemental reactions and reaction rates can be determined for the many chemical reactions present in a fluidized bed. However, the large number of coupled reactions in large-scale industrial reactors cannot be calculated within hundreds of seconds of simulation time [39–41]. The reduction reaction system between the particles and gas phase considered in this study is similar to that considered by Parisi et al. [36]. The reduction process of Fe<sub>2</sub>O<sub>3</sub> actually includes Fe<sub>2</sub>O<sub>3</sub> $\rightarrow$ Fe<sub>3</sub>O<sub>4</sub> $\rightarrow$ FeO $\rightarrow$ Fe [36]. In the simulation, the intermediate process was omitted, and the chemical reaction was directly reduced from Fe<sub>2</sub>O<sub>3</sub> to Fe. Because the water–gas shift reaction (WGSR) = reaction (3) – reaction (2), the water–gas reaction is not linearly independent and implicit in the system, and its reaction rate is not listed. The reduction of ore powder is as follows:

$$Fe_2O_3 + 3H_2 = 2Fe + 3H_2O$$
 (2)

$$Fe_2O_3 + 3CO = 2Fe + 3CO_2 \tag{3}$$

To incorporate this chemistry in CPFD, cell mean chemistry calculations were used. The homogeneous reaction rate was obtained by the Arrhenius equation, which is written as  $k = Ae^{-Ea/RT}$  [42]. The average properties of particle phases in the chemical rate equation were calculated by interpolating the discrete calculated particle properties into the grid [43–45]. The reaction rates in each grid cell were calculated by solving a set of ordinary differential equations of the above form. In the implicit solution, the mass of the solid phase involved was contained in the mass of the gas phase.For reactions (1) and (2), the consumption rates of H<sub>2</sub> and CO per unit volume are [46], respectively, as follows:

$$R_{H_2} = 3.0a_s \left[ \frac{p_{H_2} - \frac{p_{H_{2O}}}{K_{eH_2}}}{RT_S r_p} \frac{k_{H_2} (1 + K_{eH_2})}{K_{eH_2}} \right] (1 - f)^{\frac{2}{3}}$$
(4)

$$R_{CO} = 3.0a_s \left[ \frac{p_{CO} - \frac{p_{CO_2}}{K_{eCO}}}{RT_S r_p} \frac{k_{CO} (1 + K_{eCO})}{K_{eCO}} \right] (1 - f)^{\frac{2}{3}}$$
(5)

where the chemical equilibrium constants and reaction rate constants of H<sub>2</sub> and CO are, respectively, given by the following:  $K_{eH_2} = e^{\frac{-1589.6+16.21T_s}{T_s}}$ ,  $K_{H_2} = 19.5e^{\frac{-62700}{RT_s}}$ ,  $K_{eCO} = e^{\frac{22890-24.36T_s}{RT_s}}$ , and  $K_{CO} = 27.9e^{\frac{-62700}{RT_s}}$ . The generation or consumption rate of other substances in the reaction system was obtained by using the stoichiometric coefficients of reaction (Equations (2) and (3)).

#### 3.3. Simulated Experimental Conditions

According to the experimental conditions, in the simulation, the gas phase was  $N_2$ , CO, and  $H_2$ , and the solid phase was iron ore powder and silica sand. The particle size distribution of the ore powder is shown in Figure 3. The initial setup was the same as the experimental setup, as presented in Figure 4. Particles were filled above the inlet boundary with a volume fraction of 0.56, and the other input parameters are listed in Table 3. First, the temperature of the fluidized bed reactor was heated to the set value, after which  $N_2$  was introduced and the mineral powder. The gas in the experiment was introduced from the bottom of the fluidized bed to fluidize the particles and reduce the mineral powder under high-temperature conditions. The solid phase was not allowed to be discharged from the reactor, and atmospheric pressure boundary conditions were specified at the outlet. The simulation time required 2500 s to eliminate the initial oscillation and achieve complete mass dynamic balance and the mineral powder composition reaction.



Figure 3. Particle size distribution.

Table 3. Input parameters in the CPFD simulation.

Parameters	Numerical Value
Gravitational acceleration	$9.81 \text{ m/s}^2$
Diameter of mineral powder	$7.5 imes10^{-5} imes1.5 imes10^{-4}~\mathrm{m}$
Mineral powder density	$4216.81 \text{ kg/m}^3$
Adhesive diameter	$5 \times 10^{-3}$ ~ $1 \times 10^{-2}$ m
Drag model	Wen-Yu
Non-dimensional exponent, β	3
Non-dimensional constant, a	$10^{-8}$
Turbulence model	Large eddy simulation (LES) turbulence
Maximum momentum of particle collision	40%
Time step	0.001 s
Total time	2500 s



Figure 4. Initial settings.

To study the effects of the reduction temperature, the type of reducing gas, the reduction pressure, and the linear velocity of the reducing gas on the fluidized reduction of iron ore fines, experiments, and simulations were combined to investigate these factors. These four factors are, respectively, represented by A, B, C, and D. A represents the reduction temperature, and A<sub>1</sub>, A<sub>2</sub>, and A<sub>3</sub> are 923 K, 1023 K, and 1123 K, respectively. B represents the type of reducing gas, and B<sub>1</sub>, B<sub>2</sub>, and B3, respectively, represent H<sub>2</sub>, CO, and an H<sub>2</sub>:CO mixed gas with a ratio of 0.6:0.4. C represents the reduction pressure, and C<sub>1</sub>, C<sub>2</sub>, and C<sub>3</sub> represent the atmospheric pressures of 0.1 MPa, 0.2 MPa, and 0.4 MPa, respectively. Finally, D represents the linear velocity of the reducing gas, and D<sub>1</sub>, D<sub>2</sub>, and D<sub>3</sub> are, respectively, 0.4 m/s, 0.6 m/s, and 0.8 m/s (see Table 4). In the experiment, the reduction effect was analyzed by measuring the metallization rate of the ore powder, and the optimal experimental operating conditions of the fluidized bed reduction were obtained via optimization analysis.

Table 4. Levels of each factor.

A Reduction	B Reduction Gas	C Reduction	D Gas Linear
Temperature /K	Type	Pressure/MPa	Velocity/m/s
A <sub>1</sub> A <sub>2</sub> A <sub>3</sub>	$B_1 B_2 B_3 H_2 CO Mixture$	C <sub>1</sub> C <sub>2</sub> C <sub>3</sub>	D <sub>1</sub> D <sub>2</sub> D <sub>3</sub>
923 1023 1123		0.1 0.2 0.4	0.4 0.6 0.8

#### 4. Conclusion and Analysis

For different indicators, the degrees of influence of different factors are different, and the comprehensive optimal solution can be obtained via the comprehensive analysis of the four factors. Via the comprehensive analysis of the metallization rate and bonding ratio in the experiment, the optimal experimental operating conditions were determined to be a temperature of 923 K, a pressure of 0.2 MPa, a linear velocity of the reducing gas of 0.6 m/s, and the use of pure H<sub>2</sub> as the reducing gas. For the specific research process and conclusion of the experiment, please refer to the paper published by the research group [47]. Figure 5 presents the initial fluidization process of the iron ore powder. The ore powder was reduced to iron to a certain extent, and the composition remained unchanged.



Figure 5. The initial fluidization process of simulated bed.

#### 4.1. Metallization Rate

Figure 6 exhibits the changing trends of the metallization rates of the three groups of I, II, and III. Figure 6a is the simulation result of the metallization rate, and Figure 6b is the experimental measurement value of the metallization rate. By comparing the simulation data with the experimental data, it can be seen that the simulation results of the CPFD method can adequately verify the variation trend of the metallization rate observed in the experiment. Within 1800 s, the metallization rate increased with time, and after 1800 s, the metallization rate exhibited an almost stable trend and no longer changed. Since the chemical reaction rate set by the simulation is in a more ideal state, while the real experiment requires a heated reaction time to reduce the iron ore powder, the simulated value of the metallization rate in the first 600 s is in the ideal state of linear growth, while the real experimental measured value has a lower growth rate than the simulated result. Moreover, the reaction time is long enough for the powder to be mostly reduced under the simulated experiment, which leads to the metallization rate under the simulated result being larger than the metallization rate in the experimental result.



Figure 6. Metallization rate.

The experimentally measured value of the metallization rate at 1200 s was compared with the simulated calculated value, as shown in Table 5. The comprehensive analysis shows that under the same conditions, the simulated values of the metallization rate were in good agreement with the change trend of the experimentally measured values. Therefore, the simulated metallization rate can provide reference values for the optimization of the experimental operating conditions, and can be used to verify the accuracy of the experimental results. However, there was an error in the simulation result, and the calculated value was higher than the experimental value. The reason for this is that the simulation result was in an ideal state, which ignored the fact that some particles were blown away and the energy loss in the experiment. Moreover, the setting of the chemical reaction rate could not completely reflect the real situation. As the particle composition in the ideal state was without impurities, most of the mineral powder could be reduced over a sufficiently long amount of time, which caused the metal content in the simulation result to be greater than the experimentally measured value. When the metallization rate in the simulation tended to be stable, the particles under the simulation were mostly reduced, resulting in a higher metallization rate.

Table 5. Experimental scheme and results.

No.	Reduction Temperature/K	Type of Reducing Gas	Reducing Pressure/MPa	Linear Velocity/m/s	Metallization Rate /%	Simulated Metallization Rate/%
1	1123	H <sub>2</sub>	0.1	0.6	76.45	86.45
2	1023	CO	0.1	0.6	60.46	68.46
3	1023	$H_2 + CO$	0.1	0.6	63.15	70.15
4	923	$H_2 + CO$	0.2	0.6	80.04	99.13
5	923	$H_2 + CO$	0.4	0.6	81.65	99.81
6	1023	$H_2 + CO$	0.1	0.4	51.88	58.88
7	1023	$H_2 + CO$	0.1	0.8	60.19	69.19
8	1023	H <sub>2</sub>	0.2	0.4	56.87	62.87

# 4.2. Reduction Temperature

Factor A (reduction temperature): The initial temperatures of the gas and solid phases, the inlet temperature, and the wall heating temperature were simulated according to the experimental conditions, and the fluidization effect was judged by the time required for the reaction to reach equilibrium. Table 6 shows the simulation experimental scheme and results to study the reduction temperature. Figure 7 presents the fluid temperature distribution of simulation experiments 1, 2, and 3, and the cross-section temperature field distribution of simulation experiments 2. The superimposition on the right is the change of the average temperature of simulation experiments 1, 2, and 3 over time. Figure 8 shows the changes in the compositions of the three groups of experimental gases with time, which reveals that the time required for the ore powder to be completely reduced to iron to reach the equilibrium state in these experiments was approximately 1215 s, 990 s, and 1240 s. When gases of different compositions are blown into the fluidized bed from the inlet, the mineral powder is continuously reduced to Fe by the reducing gas, and when the mineral powder is completely reduced, no chemical reaction occurs in the fluidized bed, and the gas in the fluidized bed is detected as the composition set by the initial conditions. When the temperature exceeded a certain value, the higher the temperature, the poorer the reduction effect of the ore powder as compared to that at a low temperature. According to the simulation results, the shortest time required to reach the equilibrium occurred when the reduction temperature was 1023 K. When the mineral powder with a lower temperature was added to the bed, it took a period of time to increase the temperature, and the reduction effect of the mineral powder gradually increased with the reduction reaction. When the temperature reached a certain level, the reduction effect was optimal. The higher the energy of iron whiskers produced by reduction on the particle surface, the greater the physical adsorption, and the more likely the agglomeration between high-energy iron whiskers, resulting in bonding/loss of flow and a decrease in the metallization rate [47]. Therefore, the best choice for the reduction temperature was determined to be 1023 K. The simulation conclusion is consistent with the experimental conclusion, so this method can simulate the effect of the reduction temperature on fluidized ironmaking.

No.	Reduction Temperature/K	Type of Reducing Gas	Reducing Pressure/MPa	Linear Velocity/m/s	Simulation Equilibration Time/s
1	923	H <sub>2</sub>	0.2	0.6	1215
2	1023	$H_2$	0.2	0.6	990
3	1123	$H_2$	0.2	0.6	1240
4	923	$H_2$	0.1	0.6	1255
5	1023	$H_2$	0.1	0.6	1105
6	1123	H <sub>2</sub>	0.1	0.6	1305

Table 6. Experimental scheme and results.



Figure 7. Temperature distribution of fluid.



Figure 8. Changes in gas composition over time. (a) Experiments 1. (b) Experiments 2. (c) Experiments 3.

#### 4.3. Types of Reducing Gases

Factor B (type of reducing gas): Different components of the reducing gas have a great influence on the reduction effect. Table 7 shows the simulation experimental scheme and results of studying the components of the reducing gas, and Figure 9 presents the changes in the simulation results of the gas components in simulation experiments 7, 8, and 9 over time. In the three experiments, the time required for the ore particles to be completely reduced to the equilibrium state was roughly 1375 s, 1675 s, and 1580 s, respectively. The simulation results show that the higher the  $H_2$ , the shorter the equilibrium time, and the better the fluidization reduction effect. Therefore, pure  $H_2$  was determined to be the best reducing gas. When pure  $H_2$  was used as the reducing gas in the experiment, the reduction effect was good and the metallization rate was high. According to the preceding analysis, when the temperature exceeds a certain value, the lower the temperature, the more conducive the reaction. The CO reduction of iron ore is an exothermic reaction, while the H<sub>2</sub> reduction of iron ore is an endothermic reaction. When a certain temperature is reached, the high temperature is conducive to particle bonding/loss of flow, and CO will precipitate carbon powder and adsorb around the ore powder, thus affecting the reaction. The chemical reaction rate settings in the simulation were consistent with the real experimental results. This method can simulate the effect of the reducing gas on fluidized smelting.

No.	Reduction Temperature/K	Type of Reducing Gas	Reducing Pressure/MPa	Linear Velocity/m/s	Simulation Equilibration Time/s
7	1023	$H_2$	0.1	0.6	1375
8	1023	CO	0.1	0.6	1675
9	1023	$H_2 + CO$	0.1	0.6	1580
10	1023	$H_2$	0.2	0.6	1050
11	1023	CŌ	0.2	0.6	1305
12	1023	$H_2 + CO$	0.2	0.6	1155



Figure 9. Changes in gas composition over time. (a) Experiments 7. (b) Experiments 8. (c) Experiments 9.

#### 4.4. Reduction Pressure

Factor C (reduction pressure): Table 8 shows the experimental plan and results of the reduction pressure study, and Figure 10 presents the fluid temperature and particle temperature distributions of simulation experiments 13, 14, and 15. The overlay on the right shows the average temperatures of the three groups of experiments. Figure 11 exhibits the simulation results of gas changes in the three groups of experiments. It can be seen from the figure that the time required for the three groups of reactions to reach equilibrium was roughly 1625 s, 1115 s, and 1050 s, respectively. According to the experimental analysis, it can be seen that when the pressure was higher, the gas density in the bed was higher, and the contact between the gas and the solid was more sufficient, which accelerated the reduction rate of the solid. However, the higher the pressure, the higher the gas velocity. Before the set heating temperature was reached, the gas flowed out from the gas outlet, thereby reducing both the fluid temperature and the particle temperature in the fluidized bed. The higher the outlet pressure, the lower the temperature, and the better the reduction effect. However, when the pressure was 0.2 MPa and 0.4 MPa, the time required for the balance of the ore powder was very similar. Considering the cost and the reduction effect, the best reduction pressure was selected as 0.2 MPa. This method can simulate the effect of the reduction pressure on fluidized ironmaking.

No.	Reduction Temperature/K	Type of Reducing Gas	Reducing Pressure/MPa	Linear Velocity/m/s	Simulation Equilibration Time/s
13	1023	$H_2 + CO$	0.1	0.6	1625
14	1023	$H_2 + CO$	0.2	0.6	1115
15	1023	$H_2 + CO$	0.4	0.6	1050
16	923	$H_2 + CO$	0.1	0.6	1605
17	923	$H_2 + CO$	0.2	0.6	1105
18	923	$H_2 + CO$	0.4	0.6	1055

Table 8. Experimental scheme and results.



Figure 10. Temperature of fluid and particles.



Figure 11. Changes in gas composition over time. (a) Experiments 13. (b) Experiments 14. (c) Experiments 15.

#### 4.5. Linear Gas Velocity

Factor D (linear gas velocity): Table 9 shows the experimental plan and results of the Linear Gas Velocity study, and Figure 12 shows the distributions of the fluid temperature and particle temperature in simulation experiments 19, 20, and 21, as well as the crosssectional distribution of the fluid temperature in simulation experiment 20. Figure 13 presents the variations of the gas composition in the three groups of experimental beds. It can be seen from Figures 12 and 13 that when the gas velocity was 0.4 m/s, the ore powder was not completely reduced, and the gas velocity was 0.6 m/s and 0.8 m/s. When the gas velocity was 0.6 m/s, the ore powder was completely reduced and the reaction reached the equilibrium state after 1580 s, whereas when the gas velocity was 0.8 m/s, the reaction required 1875 s. Therefore, the best gas velocity was determined to be 0.6 m/s. With the increase of the linear gas velocity, the upward drag force of the gas on the particles also increases, the load of the particles decreases, the spacing of the particles increases, the tightness of the particles in the bed decreases, the porosity increases, the gas and particles fully contact and react, and adhesion/loss of flow does not easily occur. Furthermore, with the increase of the linear gas velocity, the temperature of normal-temperature gas decreases, the particle temperature and cell fluid temperature decrease, and the effective gas-solid reduction temperature and reduction effect decrease. A large temperature decrease in a

fluidized bed is not conducive to the reduction of ore powder. Considering the optimal reduction linear velocity of 0.6 m/s, it can be concluded that the method can simulate the influence of the linear gas velocity on fluidized smelting.

No.	Reduction Tempera- ture/K	Type of Reducing Gas	Reducing Pres- sure/MPa	Linear Velocity/m/s	Simulation Equilibra- tion Time/s
19	1023	$H_2 + CO$	0.1	0.4	
20	1023	$H_2 + CO$	0.1	0.6	1580
21	1023	$H_2 + CO$	0.1	0.8	1875
22	1023	H <sub>2</sub>	0.2	0.4	
23	1023	$H_2$	0.2	0.6	1055
24	1023	$H_2$	0.2	0.8	1400

Table 9. Experimental scheme and results.



Figure 12. Fluid and particle temperature distributions.



Figure 13. Changes in gas composition over time. (a) Experiments 19. (b) Experiments 20. (c) Experiments 21.

## 5. Conclusions

In this study, a combination of numerical simulation and experimental testing was used to carry out numerical simulation research on the thermal state of reduced iron ore powder in a pressurized fluidized bed with the help of Barracuda software. Coupling the characteristics of gas–solid flow and the reaction kinetics, a numerical model of the reduction of iron ore fines in a pressurized circulating fluidized bed was established to reveal the flow and reaction characteristics of the fluidized reduction of iron ore fines. The effects of the operating parameters, namely the reduction pressure, reduction temperature, reducing gas type, and linear gas velocity, on the gasification results were explored. The simulation results reveal the following:

(1) The change trends of the metallization rate in the fluidized reduction of iron ore powder simulated by the CPFD method were consistent with the experimental values;

(2) The CPFD method simulated the effects of the reduction temperature, linear gas velocity, gas composition, and reduction pressure on the results of the fluidized reduction of iron powder, which were consistent with the experimental results. This indicates that the model selection was reasonable, and lays a foundation for further numerical simulation work;

(3) Based on the CPFD method, the optimal operating parameters were determined to be a reduction temperature of 1023 K, a reduction pressure of 0.2 MPa,  $H_2$  as the reducing gas, and a gas velocity of 0.6 m/s. The optimal operating parameters in the experimental test were a reduction temperature of 1023 K, a reduction pressure of 0.2 MPa,  $H_2$  as the reducing gas, and a gas velocity of 0.6 m/s. The simulation conclusion is consistent with the experimental test conclusion, which verifies the reliability of the experimental data.

**Author Contributions:** Conceptualization, Z.-w.W. and Q.-y.X.; methodology, Z.-w.W.; software, Z.-w.W.; validation, Z.-w.W. and J.-y.H.; formal analysis, J.-y.H.; investigation, J.-y.H.; resources, Z.-w.W. and G.-m.Z.; data curation, J.-y.H.; writing—original draft preparation, J.-y.H.; writing—review and editing, J.-y.H.; supervision, J.-y.H. The two authors of Z.-w.W. and J.-y.H. made substantial contributions to the manuscript. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research was funded by the National Natural Science Foundation of China–Xinjiang Joint Fund grant number U2003124, and the National Natural Science Foundation of China grant number 51974001.

Conflicts of Interest: The authors declare no conflict of interest.

# Nomenclature

- a Pre-exponential factor
- *A* Particle acceleration
- $\alpha_g, \alpha_s$  Gas and solid phase volume fraction
- C<sub>v</sub> Specific heat capacity
- $d_p$  Particle size, m
- Dg Gas phase turbulent diffusion coefficient
- Ea Apparent activation energy
- F Force source, N
- *F<sub>s</sub>* Frictional stress between particles
- g Gravitational acceleration, m/s<sup>2</sup>
- $h_{fw}$  Local fluid wall heat transfer coefficient
- $h_p$  Dilute transfer heat coefficient
- *hg* Enthalpy of mixture
- $k_f$  Thermal conductivity of a fluid
- k Rate constant
- L Bubble hole length, m
- m Particle mass, kg
- $\delta m_{g,i}^{\cdot}$  Mass change of gas component i produced by homogeneous reaction
- $\delta m_s^{\cdot}$  Change in gas mass per unit volume, kg/m<sup>3</sup>
- $\delta m_{s,i}$  Mass transfer of gas component i from a heterogeneous reaction
- $\rho_g, \rho_s$  Gas and solid phase density, kg/m<sup>3</sup>
- *P* Average pressure, Pa
- *P<sub>r</sub>* Prandtl number
- *q* Energy transfer between gas phase and solid phase
- $q \dot{}_D$  Energy changes due to component diffusion
- $\dot{Q}$  Heat of reaction, J
- *Q<sub>ij</sub>* Thermal conductivity between particles
- *Q<sub>radi</sub>* Radiation heat transfer between particles and walls
- *Q<sub>reac</sub>* Heat of reaction
- *Q<sub>sg</sub>* Convective heat transfer between particles and gases
- R Molar gas constant
- Re Reynolds number
- $S_h$  Energy transfer between gas phase and solid phase
- *T* Thermodynamic temperature
- $V_P$  Particle volume, m<sup>3</sup>
- $u_g, u_s$  Gas and solid phase velocity, m/s
- $Y_{g,i}$  Mass fraction of gas components
- $\tau_g$  Gas phase stress tensor
- $\tau_s$  Particle collision stress
- $\varphi$  Viscous dissipation
- $\varepsilon_s$  Volume fraction of particles in unit grid
- $\theta_p$  Wall particle volume fraction
- $\theta_{cp}$  Dense packing value fraction

#### References

- 1. Kinaci, M.E.; Lichtenegger, T.; Schneiderbauer, S. A CFD-DEM Model for the Simulation of Direct Reduction of Iron-Ore in Fluidized Beds. *Chem. Eng. Sci.* 2020, 227, 115858. [CrossRef]
- Guenther, C.; Syamlal, M. The Effect of Numerical Diffusion on Simulation of Isolated Bubbles in a Gas–Solid Fluidized Bed. Powder Technol. 2001, 116, 142–154. [CrossRef]
- 3. Kunii, D.; Levenspiel, O. Fluidization Engineering; Butterworth-Heinemann: Boston, MA, USA, 1991; ISBN 0-409-90233-0.
- 4. Olofsson, G.; Ye, Z.; Bjerle, I.; Andersson, A. Bed Agglomeration Problems in Fluidized-Bed Biomass Combustion. *Ind. Eng. Chem. Res.* **2002**, *41*, 2888–2894. [CrossRef]
- 5. Chladek, J.; Jayarathna, C.K.; Moldestad, B.M.E.; Tokheim, L.-A. Fluidized Bed Classification of Particles of Different Size and Density. *Chem. Eng. Sci.* 2018, 177, 151–162. [CrossRef]
- 6. Jayarathna, C.K.; Halvorsen, B.M.; Tokheim, L.-A. Experimental and Theoretical Study of Minimum Fluidization Velocity and Void Fraction of a Limestone Based CO<sub>2</sub> Sorbent. *Energy Procedia* **2014**, *63*, 1432–1445. [CrossRef]

- Rodríguez, J.M.; Sánchez, J.R.; Alvaro, A.; Florea, D.F.; Estévez, A.M. Fluidization and Elutriation of Iron Oxide Particles. A Study of Attrition and Agglomeration Processes in Fluidized Beds. *Powder Technol.* 2000, 111, 218–230. [CrossRef]
- 8. Marco, E.; Santos, A.; Menéndez, M.; Santamaría, J. Fluidization of Agglomerating Particles: Influence of the Gas Temperature and Composition on the Fluidization of a Li/MgO Catalyst. *Powder Technol.* **1997**, *92*, 47–52. [CrossRef]
- Kraft, S.; Kirnbauer, F.; Hofbauer, H. CPFD Simulations of an Industrial-Sized Dual Fluidized Bed Steam Gasification System of Biomass with 8 MW Fuel Input. Appl. Energy 2017, 190, 408–420. [CrossRef]
- O'Rourke, P.J.; Zhao, P.; Snider, D. A Model for Collisional Exchange in Gas/Liquid/Solid Fluidized Beds. *Chem. Eng. Sci.* 2009, 64, 1784–1797. [CrossRef]
- Abbasi, A.; Ege, P.E.; de Lasa, H.I. CPFD Simulation of a Fast Fluidized Bed Steam Coal Gasifier Feeding Section. *Chem. Eng. J.* 2011, 174, 341–350. [CrossRef]
- 12. Jia, C.; Li, J.; Chen, J.; Cui, S.; Liu, H.; Wang, Q. Simulation and Prediction of Co-Combustion of Oil Shale Retorting Solid Waste and Cornstalk in Circulating Fluidized Bed Using CPFD Method. *Appl. Therm. Eng.* **2020**, *165*, 113574. [CrossRef]
- Atsonios, K.; Nikolopoulos, A.; Karellas, S.; Nikolopoulos, N.; Grammelis, P.; Kakaras, E. Numerical Investigation of the Grid Spatial Resolution and the Anisotropic Character of EMMS in CFB Multiphase Flow. *Chem. Eng. Sci.* 2011, 66, 3979–3990. [CrossRef]
- 14. Zeneli, M.; Nikolopoulos, A.; Nikolopoulos, N.; Grammelis, P.; Kakaras, E. Application of an Advanced Coupled EMMS-TFM Model to a Pilot Scale CFB Carbonator. *Chem. Eng. Sci.* **2015**, *138*, 482–498. [CrossRef]
- 15. Zhang, N.; Lu, B.; Wang, W.; Li, J. Virtual Experimentation through 3D Full-Loop Simulation of a Circulating Fluidized Bed. *Particuology* **2008**, *6*, 529–539. [CrossRef]
- Zhang, N.; Lu, B.; Wang, W.; Li, J. 3D CFD Simulation of Hydrodynamics of a 150 MWe Circulating Fluidized Bed Boiler. *Chem. Eng. J.* 2010, *162*, 821–828. [CrossRef]
- 17. Zhou, Q.; Wang, J. CFD Study of Mixing and Segregation in CFB Risers: Extension of EMMS Drag Model to Binary Gas–Solid Flow. *Chem. Eng. Sci.* **2015**, *122*, 637–651. [CrossRef]
- 18. Bidwe, A.R.; Duelli (Varela), G.; Dieter, H.; Scheffknecht, G. Experimental Study of the Effect of Friction Phenomena on Actual and Calculated Inventory in a Small-Scale CFB Riser. *Particuology* **2015**, *21*, 41–47. [CrossRef]
- Rehling, B.; Hofbauer, H.; Rauch, R.; Aichernig, C. BioSNG—Process Simulation and Comparison with First Results from a 1-MW Demonstration Plant. *Biomass Conv. Bioref.* 2011, 1, 111–119. [CrossRef]
- 20. Barata, J. Modelling of Biofuel Droplets Dispersion and Evaporation. Renew. Energy 2008, 33, 769–779. [CrossRef]
- Liang, Y.; Zhang, Y.; Li, T.; Lu, C. A Critical Validation Study on CPFD Model in Simulating Gas–Solid Bubbling Fluidized Beds. *Powder Technol.* 2014, 263, 121–134. [CrossRef]
- Lim, J.-H.; Bae, K.; Shin, J.-H.; Kim, J.-H.; Lee, D.-H.; Han, J.-H.; Lee, D.H. Effect of Particle–Particle Interaction on the Bed Pressure Drop and Bubble Flow by Computational Particle-Fluid Dynamics Simulation of Bubbling Fluidized Beds with Shroud Nozzle. *Powder Technol.* 2016, 288, 315–323. [CrossRef]
- 23. Zhou, Y.; Zhu, J. A Review on Fluidization of Geldart Group C Powders through Nanoparticle Modulation. *Powder Technol.* 2021, 381, 698–720. [CrossRef]
- Chu, K.W.; Wang, B.; Yu, A.B.; Vince, A. CFD-DEM Modelling of Multiphase Flow in Dense Medium Cyclones. *Powder Technol.* 2009, 193, 235–247. [CrossRef]
- 25. Bandara, J.C.; Jayarathna, C.; Thapa, R.; Nielsen, H.K.; Moldestad, B.M.E.; Eikeland, M.S. Loop Seals in Circulating Fluidized Beds—Review and Parametric Studies Using CPFD Simulation. *Chem. Eng. Sci.* **2020**, 227, 115917. [CrossRef]
- Wu, Y.-L.; Jiang, Z.-Y.; Zhang, X.-X.; Xue, Q.-G.; Miao, Z.; Zhou, Z.; Shen, Y.-S. Process Optimization of Metallurgical Dust Recycling by Direct Reduction in Rotary Hearth Furnace. *Powder Technol.* 2018, 326, 101–113. [CrossRef]
- 27. Hou, Q.-F.; Samman, M.; Li, J.; Yu, A.-B. Modeling the Gas-Solid Flow in the Reduction Shaft of COREX. *ISIJ Int.* 2014, 54, 1772–1780. [CrossRef]
- Stollhof, M.; Penthor, S.; Mayer, K.; Hofbauer, H. Influence of the Loop Seal Fluidization on the Operation of a Fluidized Bed Reactor System. *Powder Technol.* 2019, 352, 422–435. [CrossRef]
- 29. Weber, G.; Di Giuliano, A.; Rauch, R.; Hofbauer, H. Developing a Simulation Model for a Mixed Alcohol Synthesis Reactor and Validation of Experimental Data in IPSEpro. *Fuel Process. Technol.* **2016**, *141*, 167–176. [CrossRef]
- 30. Mehrabian, R.; Scharler, R.; Obernberger, I. Effects of Pyrolysis Conditions on the Heating Rate in Biomass Particles and Applicability of TGA Kinetic Parameters in Particle Thermal Conversion Modelling. *Fuel* **2012**, *93*, 567–575. [CrossRef]
- 31. Kirnbauer, F.; Hofbauer, H. Investigations on Bed Material Changes in a Dual Fluidized Bed Steam Gasification Plant in Güssing, Austria. *Energy Fuels* **2011**, *25*, 3793–3798. [CrossRef]
- 32. Snider, D.M. Three Fundamental Granular Flow Experiments and CPFD Predictions. Powder Technol. 2007, 176, 36–46. [CrossRef]
- Valipour, M.S.; Motamed Hashemi, M.Y.; Saboohi, Y. Mathematical Modeling of the Reaction in an Iron Ore Pellet Using a Mixture of Hydrogen, Water Vapor, Carbon Monoxide and Carbon Dioxide: An Isothermal Study. *Adv. Powder Technol.* 2006, 17, 277–295. [CrossRef]
- 34. Yan, R.; Liang, D.T.; Laursen, K.; Li, Y.; Tsen, L.; Tay, J.H. Formation of Bed Agglomeration in a Fluidized Multi-Waste Incinerator. *Fuel* **2003**, *82*, 843–851. [CrossRef]
- Loha, C.; Chattopadhyay, H.; Chatterjee, P.K. Three Dimensional Kinetic Modeling of Fluidized Bed Biomass Gasification. *Chem. Eng. Sci.* 2014, 109, 53–64. [CrossRef]

- 36. Parisi, D.R.; Laborde, M.A. Modeling of Counter Current Moving Bed Gas-Solid Reactor Used in Direct Reduction of Iron Ore. *Chem. Eng. J.* 2004, 104, 35–43. [CrossRef]
- 37. van Wachem, B.G.M.; Almstedt, A.E. Methods for Multiphase Computational Fluid Dynamics. *Chem. Eng. J.* **2003**, *96*, 81–98. [CrossRef]
- Hu, H.; Zhou, Q.; Zhu, S.; Meyer, B.; Krzack, S.; Chen, G. Product Distribution and Sulfur Behavior in Coal Pyrolysis. *Fuel Process. Technol.* 2004, *85*, 849–861. [CrossRef]
- 39. Chu, K.W.; Wang, B.; Xu, D.L.; Chen, Y.X.; Yu, A.B. CFD–DEM Simulation of the Gas–Solid Flow in a Cyclone Separator. *Chem. Eng. Sci.* 2011, *66*, 834–847. [CrossRef]
- Adamczyk, W.P.; Klimanek, A.; Białecki, R.A.; Węcel, G.; Kozołub, P.; Czakiert, T. Comparison of the Standard Euler–Euler and Hybrid Euler–Lagrange Approaches for Modeling Particle Transport in a Pilot-Scale Circulating Fluidized Bed. *Particuology* 2014, 15, 129–137. [CrossRef]
- 41. Benyahia, S.; Galvin, J.E. Estimation of Numerical Errors Related to Some Basic Assumptions in Discrete Particle Methods. *Ind. Eng. Chem. Res.* **2010**, *49*, 10588–10605. [CrossRef]
- Austin, P.R.; Nogami, H.; Yagi, J. A Mathematical Model for Blast Furnace Reaction Analysis Based on the Four Fluid Model. *ISIJ* Int. 1997, 37, 748–755. [CrossRef]
- 43. Negri, E.D.; Alfano, O.M.; Chiovetta, M.G. Direct Reduction of Hematite in a Moving-Bed Reactor. Analysis of the Water Gas Shift Reaction Effects on the Reactor Behavior. *Ind. Eng. Chem. Res.* **1991**, *30*, 474–482. [CrossRef]
- 44. Usui, T.; Ohmi, M.; Yamamura, E. Analysis of Rate of Hydrogen Reduction of Porous Wustite Pellets Basing on Zone-Reaction Models. *ISIJ Int.* **1990**, *30*, 347–355. [CrossRef]
- 45. Nakhaei, M.; Wu, H.; Grévain, D.; Jensen, L.S.; Glarborg, P.; Dam–Johansen, K. CPFD Simulation of Petcoke and SRF Co–Firing in a Full–Scale Cement Calciner. *Fuel Process. Technol.* **2019**, *196*, 106153. [CrossRef]
- 46. Wang, L.X. Simulation Study of a New Smelting Reduction System Based on Iron Fine Ore; Chinese Academy of Sciences: Beijing, China, 2000.
- Xu, Q.; Gu, Z.; Wan, Z.; Huangfu, M.; Meng, Q.; Liao, Z.; Wu, B. Effect of Coated Cow Dung on Fluidization Reduction of Fine Iron Ore Particles. *Processes* 2021, 9, 1175. [CrossRef]