Effect of High Reactivity Coke for Mixed Charge in Ore Layer on Reaction Behavior of Each Particle in Blast Furnace

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A low coke rate operation in blast furnace is desired to decrease the carbon input and mitigate global warming problem. However, low coke rate operation tends to cause the gas permeability to deteriorate. The mixing of small-size coke (nut coke) including high reactivity coke in ore layer is considered to be a promising way to improve permeability and increase reaction efficiency in a blast furnace. Although adding a nut coke mixing to an ore layer is predicted to be empirically effective in low coke rate operation, there is little actual data on microscopic phenomena of each particle in the packed bed. In the present study, an Euler–Lagrange approach was introduced to precisely understand the influence of the packed bed structure on the reaction behavior of each particle in the three-dimensional particle arrangement.

It was observed that the heterogeneity on the reaction rate and temperature distribution was influenced by the particle arrangement. When high-reactivity coke was used at approximately 1273 K, although CO gas fraction increased, the gaseous phase temperature decreased due to the active solution loss reaction rate of the nut coke in the mixed layer. As a result, the ore reduction rate decreased. The contribution of high-reactivity coke to the ore reduction rate depends on the particle arrangement through the heat transfer and reaction heat. Accordingly, in the case of the mixed charge of the high reactivity nut coke in the ore layer, the design of the packed bed structure is important.

KEY WORDS: ironmaking; blast furnace; low coke rate; reducing agent; mixed coke charge; nut coke; reactivity; Euler–Lagrange method.

1. Introduction

In a blast furnace, a low coke rate operation is desired to mitigate global warming problem. Regarding the low reducing agent operation, several ideas have been proposed. Suitable application of small-size coke (nut coke) mixed charge including high-reactivity coke has received special attention because it is a promising approach to decreasing coke rate owing to the improved reaction efficiency of the blast furnace. Nomura et al. reported that high reactivity coke (e.g., ferro-coke) starts to react with CO$_2$ at a lower temperature, which lowers the temperature range of the thermal reserve zone in blast furnaces. Then, the CO/CO$_2$ ratio in the gas phase equilibrated with FeO and metallic Fe changes with a decreased temperature and the utilization ratio of carbon based on the wüstite–iron reduction equilibrium increases. Higuchi et al. confirmed that enhancing coke reactivity is effective for increasing the gas utilization ratio through the change in the thermal reserve zone temperature using BIS test furnace.

Although the stable operation of blast furnace in low reducing agent condition should be pursued, understanding in-furnace phenomena in blast furnace on the basis of non-empirical methods is becoming increasingly necessary. Because the microscopic phenomena that occur in a blast furnace are difficult to observe, mathematical models help in understanding the mechanism of such phenomena. As described above, the charge of nut coke and high reactivity coke is beneficial to attain low reducing agent operation, and the mechanism of chemical reaction and heat transfer must be clarified to design the favorable packed bed structure.

From the above backgrounds, the present study newly considers the effect of mixed different-reactivity cokes on the reduction of ore at the particle scale and provides information on the behavioral influence under each arrangement of different particles. In particular, this study focused on the particle arrangement of high reactivity coke in the packed bed. A large-scale direct simulation that is assumed to be of the scale of an experimental blast furnace was carried out. This approach can be used to analyze the heat and mass transfer through an actual packed bed in order to verify the reduction properties and flow characteristics, which depend on the particle arrangement.

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2. Mathematical Modeling

In order to understand in-furnace gas–solid transport phenomena including momentum, heat, and mass transfers, many numerical analysis models of blast furnaces have been developed. A suitable method according to the situation selected. Figure 1 is a schematic illustration showing the arrangement of calculated points in gas–solid flow analysis, which is available for this simulation. The Euler–Euler (E–E) method, which is also called the two-fluid model, permits steady-state analysis even if the analysis object is a large-scale system such as a huge blast furnace. At present, this method can predict the change in the melting zone from the temperature distribution. Although there have been considerable researches focused on burden distribution modeling, calculating the discrete inter-particle stress using the E–E method is too complicated. Using the Euler–Lagrange (E–L) method is much easier; this approach can generate transport equations of an individual solid particle. Although the E–L method has a heavy computational load, current developments in computer technology has made it possible to apply it to large-scale systems.

The authors previously developed an entire blast furnace gas–solid flow analysis model using the E–L method. The latest article discussed using the E–L method for the heat and mass transfer model and chemical reaction model. Based on this approach, the interaction between cokes and ores under fluid flow in accordance with the movement and properties of individual particles can be directly understood in three dimensions, even if high-reactivity nut coke is used. Here, the E–L method was applied to obtain the microscopic phenomena such as reaction rate and temperature for each particle in the packed bed.

3. Governing Equations

Figure 2 shows a fixed volume element with arrows to indicate the directions of the fluxes through the surfaces by all transport phenomena in the packed bed. The gas–solid phase momentum, heat, and mass transfer and exchange are described by the following equations.

3.1. Motion Equations

3.1.1. Motion of Particles

A particle’s motion depends on Newton’s second law, as shown in Fig. 2(b). Because a solid particle has a small size, the Basset term and Saffman and Magnus lift forces are neglected.

$$m_p \frac{du_p}{dt} = \sum_{j=1}^{N_c} F_{cp} + F_{d} + F_{g} \quad \text{(1)}$$
$$I_p \frac{d\omega_p}{dt} = r_p \sum_{j=1}^{N_c} (F_{cT} - F_{cT}) \quad \text{(2)}$$

where $m_p$ is the particle mass, $u_p$ is the particle velocity, $t$ is time, $F_c$ is the particle–particle contact force, $N_c$ is the contact particle number, $F_d$ is the particle–fluid interaction force, $F_g$ is the gravity force, $I_p$ is the moment of inertia, $\omega_p$ is the angular velocity, $r_p$ is the article radius, $F_{cT}$ is the normal component of the contact force, $F_{cT}$ is the tangential component of contact force, and $F_r$ is the rolling friction force. The interparticle contact force is calculated by the “soft sphere” assumption based on Hertz’s contact theory; the same approach was used in previous research.
3.1.2. Motion of a Fluid

The movement of a fluid is described by the continuity equation and incompressible Navier–Stokes equation, which neglect the Reynolds stress:

$$\frac{\partial}{\partial t} (\rho \mathbf{u}) + \nabla \cdot (\rho \mathbf{u} \mathbf{u}) = 0 \quad \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots 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where the overall reaction rate coefficient \( k_m \) is given from the assumed compound rate of the chemical reaction and mass transfer:

\[
k_m = \left( \frac{1}{k_d} + \frac{1}{\eta c_{l}} \right)^{-1}
\]

where \( k_d \) is the mass transfer coefficient, \( k_c_{l} \) is the chemical reaction rate coefficient of equation \( l \), and \( \eta c_{l} \) is the catalytic effectiveness factor.

To determine \( k_d \), Wakao’s equation \(^{30}\) is applied in the low void fraction region (\( \varepsilon < 0.8 \)), and Usui’s equation \(^{31}\) is applied in the high void fraction region (\( \varepsilon > 0.8 \)). \( \eta c_{l} \) is given by the function of the Thiele number, which is the ratio of the ideal reaction rate. Diffusion in the grain was assumed to be negligible, and the inside of a particle was assumed to have the same temperature and composition as the surface. \( K_c_{l} \) is given from each reaction equation \( l \).

(a) Solution loss reaction \( l = 1 \)

\[
C(s) + \text{CO}_2(g) = 2\text{CO}(g)
\]

(b) Reduction reaction of hematite by CO \( l = 2 \)

\[
3\text{Fe}_2\text{O}_3(s) + \text{CO}(g) = 2\text{Fe}_3\text{O}_4(s) + \text{CO}_2(g)
\]

(c) Reduction reaction of magnetite by CO \( l = 3 \)

\[
\text{Fe}_3\text{O}_4(s) + \text{CO}(g) = 3\text{FeO}(s) + \text{CO}_2(g)
\]

(d) Reduction reaction of wüstite by CO \( l = 4 \)

\[
\text{FeO}(s) + \text{CO}(g) = \text{Fe}(s) + \text{CO}_2(g)
\]

3.5. Reaction Heat

The reaction heat is calculated from Kirchhoff’s law, which relates enthalpy to the composition and temperature.

\[
\Delta H_{r,l} = \Delta H_{298,K,l} + \int_{298K}^{T_r} C_p dT
\]
Here, ΔHform,i is the standard enthalpy change of formation, and ΔCp,i is the heat capacity change of chemical equation. l. ΔHform,i; the heat capacity coefficients, and the heats of formation are from Ref. 36.

3.6. Physical Properties
3.6.1. Gas Properties

ρg is calculated from the ideal gas equation; thus, it is a function of temperature and pressure. μg and Δg are calculated from the kinetic theory of molecules using component properties calculated from the Chapman–Enskog equation.39,40

3.6.2. Solid Properties

The ρs,i of coke is assumed to be constant, and the ρs,i of ore is calculated from the overall reduction degree of iron oxide. The particle shape is a complete sphere; thus, the particle is represented by its particle diameter. The dp of coke decreases relative to \( \frac{1}{3} \psi_s \) based on the carbon mass fraction using the shrinking core model,25 and the dp of ore is assumed to be constant. In this study, each particle size was set as much as possible close to actual particle diameter. \( \lambda_p \) is constant. The Young’s modulus and Poisson’s ratio are the same as specified in the previous article.12

3.7. Solution Method

The particle’s new velocity and position after time step \( \Delta t \) are calculated explicitly. The second-order Adams–Bashforth method was used for time marching, and the Crank–Nicholson method was used for particle movement. The distinct cell model was used for high-speed particle detection.41 For fluids, the well-known Semi-Implicit Method for Pressure-Linked Equations (SIMPLE) scheme42 was used to solve Eqs. (3), (4), (8), and (12) and the conservation of momentum, energy, and mass for a fluid cell. Each term was discretized with second-order accuracy. The local void fractions for a fluid cell were obtained from the particle locations.12 Original programs written in Fortran90/95 were used in this study; these were compiled by the Intel Fortran compiler Ver. 11.1 on UNIX and then run. In order to reduce the computation time, parallel processing of the program code was performed by OpenMP because over 2,000,000 particles were treated in this research. The CPU was an Intel Xeon® X5680 (3.33 GHz, 6 cores) × 2, and the amount of memory was 24 GB.

3.8. Calculation Conditions

The simulated packed bed and boundary conditions are illustrated in Fig. 3. If the blast furnace is considered to have a total inner volume of approximately 5,000 m³, a huge number of particles must be treated. In order to reduce the computation load, the calculation domain was set to a 2.0 m radius and 12.0 m height and assumed to be the shaft part of a blast furnace. The initially packed bed structure was derived by calculation of the particle charging processes. Coke and ore particles were alternately charged, and the initial conditions were formed as a layered structure. Our interest was in the influence of nut coke on the reduction behavior of ores when it is mixed in an ore layer. In earlier studies, two types of initial conditions were considered to determine the influence of particle arrangement: (a) coke–mix–ore (C–M–O) and (b) coke–ore–mix (C–O–M).43,44 These studies were at the laboratory scale, however; small experiments included several scale factors, and they sometimes emphasized differences from the actual scale. A large-scale direct simulation assumed to be the scale of a test blast furnace was carried out. In this study, the total particle number was 2,373,990, and the particle number ratio of coke and ore was kept to 1:1 at the mixed layer. The diameter of nut cokes particles was set to the same value of ores in order to avoid percolation. The layer thickness ratio between coke, mix, and ore was kept at 1:1:2 under the assumption of a low coke condition. Hence, the ore layer was approximately 1.2 m thick, and the coke and mix layers were approximately 0.6 m thick each. Under the assumption of a low amount of reducing agent, the coke rate was set to 249.3 kg/t, and the ore-to-coke (O/C) weight ratio was 6.37.

The calculation conditions are shown in Table 1. The

| Table 1. Calculation conditions. |
|--------------------------------|
| Solid parameters | coke | nut coke | ore |
| Particle number, – | 98 100 | 351 279 | 1 924 611 |
| Particle diameter, m | 0.080 | 0.040 | 0.040 |
| Young’s modulus, Gpa | 5.40 | 5.40 | 35.00 |
| Density, kg/m³ | 1 050 | 1 050 | 2 220 |
| Friction coefficient, – | 0.43 | 0.43 | 0.43 |
| Initial temperature, K | 1 253 | 1 253 | 1 253 |
| Heat conductivity, kW/m·K | 0.005 | 0.005 | 0.005 |
| Initial solid compositions | | | |
| mass% C | 88.1 | 88.1 | 0.0 |
| mass% Fe₂O₃ | 0.0 | 0.0 | 79.7 |
| mass% FeO₂ | 0.0 | 0.0 | 0.0 |
| mass% FeO | 0.0 | 0.0 | 5.4 |
| mass% Fe | 0.0 | 0.0 | 0.0 |
| mass% Al₂O₃ | 3.2 | 3.2 | 1.9 |
| mass% CaO | 0.004 | 0.004 | 7.2 |
| mass% SiO₂ | 8.3 | 8.3 | 4.8 |
| mass% MgO | 0.0 | 0.0 | 0.008 |
| Gas parameters | mix gas | | |
| Number of grids | 60×10×60 | | |
| Cell size | 0.20000 | | |
| Inlet pressure, kPa | 101 325 | | |
| Initial temperature, K | 1 253 | | |
| Time step, s | 1.0×10⁻¹⁰ | | |
| Inlet gas compositions | | | |
| vol% CO | 30.0 | | |
| vol% CO₂ | 10.0 | | |
| vol% H₂ | 0.0 | | |
| vol% H₂O | 0.0 | | |
| vol% N₂ | 60.0 | | |
| vol% O₂ | 0.0 | | |
inlet gaseous phase conditions assumed a bosh gas without moisture. In order to analyze the temperature effect on the reduction behavior, the inflow temperature $T_{in}$ was set to 1 273 K, and the initial temperature of the system was $(T_{in}−20)$ K. The calculation was carried out up to $t = 3 600$ s in consideration of blast furnace operation condition and computational load. The reaction rate of nut coke was based on Ref. 27).

4. Results and Discussions

4.1. Distribution of CO Fraction and Gas Temperature

First, we consider the effect of nut coke on the gas components and temperature distribution. Figure 4 shows the vertical cross-sectional distributions of the CO volume fraction which passes along the calculation domain center under each condition at $t = 3 600$ s. Note that the range of this scale is from 0.26 to 0.30. The CO fraction was gradually reduced with repeated increases and decreases through repetition of the reduction and solution loss reactions. The change in gas composition was observed at the boundary between the coke and ore layers where a mass flow and different reactions coexisted. The CO fraction was approximately 2% higher in the domain of three particles from a wall surface. This is because restraining the packed particles with the wall changes the structure; this reduces the retention time of the gaseous phase in the packed bed, so the gas is rich in CO.14) The inclined layer shape was also reflected by the CO fraction in the radial direction. In case of a) and b), CO fraction at the top of the packed bed was slightly increased by the use of high reactivity coke due to the increase in solution loss reaction rate, as represented in Eq. (17). If the nut coke reaction rate is same to lump coke, there is no remarkable difference in particle arrangement conditions between C–M–O and C–O–M except for the inner unit.

Figures 5 and 6 shows the longitudinal gas temperature and CO fraction distribution at $t = 3 600$ s, respectively. The temperature fluctuated with the same period as the CO fraction corresponding to the particle arrangement. The exothermic reaction of ore reduction and the endothermic reaction of coke solution loss appeared reciprocally. In case of high reactivity coke of a) and c), the temperature fluctuation became remarkable due to the intensified solution loss reaction of coke. At this time, almost the entire calculation domain reached a steady state. Therefore, this model was in a similar state as the thermal reserve zone in a blast furnace.

Figure 5(a) shows that the gas temperature periodically varied greatly with the high endothermic reaction of high-reactivity coke. The gas temperature in the thermal reserve zone stayed around 1 273 K when conventional coke was used; however, it decreased to 1 240 K when high-reactivity coke was applied. The thermal reserve zone temperature has been reported to show a decreasing tendency with increased coke reactivity by the mathematical model simulations.28) In this study, however, the coke rate was set very low to 249.3 kg/t; thus, a remarkable temperature increase was seen due to heat from the reduction reaction. Part of the ore layer kept the initial temperature, however the temperature in the lower part of each ore layer was reduced by the heat exchange with the coke. Because the reaction heat was divided by the ratio of specific heat between the solid and gas, it mainly contributed to the temperature change of the solid. This greatly influenced the particle reaction rate, which is described in the following section.
4.2. Reduction Degree Distribution for Each Ore Particle

A detailed investigation was carried out on the ore reduction rate. Figure 7 shows the vertical cross-sectional distributions of the degree of ore reduction passing along the calculation domain center in each condition at \( t = 3600 \) s. Note that the coke particles are indicated in blue. Most particles have not moved from initial arrangement. A remarkable increase appeared near the wall due to the above mentioned wall effect. Here, the reduction degree of ore in the mixed layer was lower than that in the ore layer. Moreover, the reduction degree of the ore layer became

![Figure 7. Ore particle reduction degree distributions. Coke particles are indicated by blue particles (\( t = 3600 \) s, C: coke, O: ore, M: mix).](image)

![Figure 8. Ore particle reduction degree distributions at vertical cross section of 2.0 m < h < 4.5 m at \( t = 3600 \) s. Coke particles are indicated by blue particles.](image)

![Figure 9. Coke particle reaction degree distributions. Ore particles are indicated by blue particles (\( t = 3600 \) s, C: coke, O: ore, M: mix).](image)

![Figure 10. Coke particle gasification degree distributions at vertical cross section of 3.5 m < h < 5.3 m at \( t = 3600 \) s. Ore particles are indicated by blue particles.](image)
slightly lower when high-reactivity coke was used (a, c) rather than conventional coke (b, d).

In order to discuss the reduction rate for each particle, Fig. 8 shows a partially enlarged view of the distribution for the ore particle reduction degree. Note that the range of this scale on the reduction degree was narrow from 6.00 to 6.65. On this scale, the change in reduction degree was seen on the particle level. This heterogeneity was influenced by the particle arrangement. The reduction rate in the upper part of the ore layers was increased by the relatively high temperature; however, the upper ore showed a lower reduction rate than the mixed layer.

When conventional coke was used (b, d), the change in reduction degree due to the arrangement was not remarkable. However, the particle arrangement greatly influenced the reduction degree when high reactivity coke was used (a, c). The reduction degree for an ore layer was higher for the arrangement shown in (c) than that shown in (a). Such a change in ore reduction degree would directly affect the behavior of the cohesive zone. Because the melting point of oxide and pig iron are influenced by the concentrations of iron oxide and carbon, respectively, the oxygen potential is correlated to liquid forming.\(^4\) In the arrangement shown in (c), the solid–liquid coexistence cohesive region shifted to the lower part of the blast furnace due to the high reduction degree of the ore layer. On the other hand, it was estimated that ores with a low reduction degree in the mixed layer extend the solid–liquid coexistence cohesive region.

A comparison of Figs. 5, 6, 7 and 8 shows that because reaction heat is transported to the upper part mostly by convective heat transfer, the large endothermic reaction of high reactivity coke makes the temperature of the layer just above significantly lower. Thus, using high reactivity coke does not enhance ore reduction reaction. At 1 273 K, the decrease in temperature becomes a more dominant factor with regard to the decrease in reduction degree than the increase in the CO fraction. Yokoyama et al.\(^4\) carried out a laboratory-scale reduction experiment on an ore particle bed and reported that the reduction degree increased when high reactivity coke was uniformly mixed in an ore layer or arranged to be in the lower part of the ore layer. This is because their conditions involved a high coke rate of O/C = 4.6 and a temperature that increased continuously from 1 173 K to 1 473 K. The change in ore reducing rate by the use of high-reactivity coke is influenced by the coke rate, and the thermal reserve zone temperature is influenced by the use of high reactivity coke. In this study, although the charge of high reactivity coke has a negative effect on the reduction rate of ore due to the intensified solution loss reaction, these phenomena could be influenced by the reducibility of ore. When high reducibility ore was used, the reduction degree would be improved owing to the change of gas composition corresponding to Fe/FeO equilibrium.

### 4.3. Gasification Degree Distribution for Each Coke Particle

Figure 9 shows the vertical cross-sectional distributions of the degree of coke reaction in each condition at t = 3 600 s, and Fig. 10 shows a partially enlarged view of the distribution for the coke particle gasification degree. Ores are indicated as blue particles. As shown in (c), the gasification degree of lump coke notably decreased compared with the other conditions. This is because the lump coke was arranged just above a mixed layer including a considerable endothermic reaction from the high-reactivity cokes, so the temperature of the coke layer was decreased. The arrangement of (c) could keep the permeability in the lower part of the furnace by protecting the lump coke so that it keeps its strength and grain size. As described in the preceding section, it might also improve ore reduction. When using high-reactivity coke, it should be just above an ore layer and directly under a coke layer (C–O–M).

Figure 11 shows the time change of the average gasification degree when high-reactivity coke was used. Both arrangement conditions indicated similar gasification rates for nut coke. However, when lump coke was compared, (a) showed a linear increase with time, but (c) showed a decrease in the reaction rate with time. Based on this result, high-reactivity coke gasifies more preferentially than lump coke in (c). By protecting lump cokes, the permeability might be maintained through keeping coke slits in the melting zone.

#### 4.4. Consideration of Reaction Heat Transfer

In this study, the particle diameter was the smallest unit of spatial discretization for calculation considered for heat and mass transfer. As we noted in our last article, the reaction heat is the dominant issue for heat transfer of each par-
ticle at this temperature scale. Therefore, in this research, the reaction heat generated on the solid surface was divided between the solid and gas phases; understanding its mechanism is important to controlling temperature distribution. This is understood at spatial scales below the particle diameter. We assumed that the particle temperature is constant, so \( \frac{1}{\kappa_p} = \infty \). The reaction heat on the solid surface is transferred to the inside of particle instantly. The quantity of reaction heat that accumulates inside the particles may have been overestimated.

One of our interests is the temperature distribution inside a solid particle and at its surface. In this case, if the surface imagery can be secured by a direct numerical simulation method, as shown in Fig. 1(c), the surface temperature distribution can be considered without too much trouble. Further examination will be required for the reaction mechanism and its heat distribution at the surface between a solid and gas.

5. Conclusions

The heat and mass transport behavior through a packed bed was studied in order to verify that the reduction properties and flow characteristics depend on the particle arrangement. In particular, the influence of small-size coke was considered for low reducing agent operation. In order to analyze the discontinuous behavior of each particle, a three-dimensional packed bed analysis model based on the Euler–Lagrange approach was used to reflect discrete particle properties; the model included the simultaneous analysis of particle and fluid flow. From the calculated results, the following results were obtained.

The exothermic reaction of ore reduction and the endothermic reaction of coke solution loss influenced each other in the packed bed. Therefore, when high-reactivity coke was used, although CO fraction increased, the decrease in the gaseous phase temperature occurred due to the intensified solution loss reaction rate at the nut coke mixed layer. As a result, the ore reduction rate decreased.

When using high-reactivity coke in the order of coke–ore–mix from the bottom, the reaction rate of the ore layer became relatively higher and the reaction rate of the lump coke layer decreased. In this arrangement, high reactivity coke was preferentially gasified to lump coke. The contribution of high reactivity coke to the ore reduction rate depends on the particle arrangement through the heat transfer and reaction heat. Accordingly, in the case of the mixed charge of the high reactivity nut coke in the ore layer, the design of the packed bed structure is important.

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