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学 位 論 文 題 目	Analysis of Iron Ore Reduction Focusing on Internal Distribution in Single Particle and Packed Bed for Hydrogen Ironmaking （水素製鉄に向けた単一粒子内および充填層内分布に着目した 鉄鉱石の還元解析）
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論文内容要約

Carbon neutrality is being pursued as a solution to the global environmental crisis. Among industrial processes, the steel industry is the largest source of CO_2 emissions. The ironmaking process relies on carbon-based gases, such as CO, as reductants to reduce iron ores, including sintered ore, lump ore, and pellets. Therefore, the different characteristics of these iron ores, such as particle shape, lead to variations in the reduction behavior. These variations in particle shape significantly impact the reduction behavior, as the reduction reactions depend on the diffusion of reductant gases and their chemical reactions. To achieve carbon neutrality, hydrogen ironmaking process has been considered as a means to reduce CO_2 emissions by replacing CO with H_2 as the reductant gas. This H_2-based reduction results in the emission of H_2O gas instead of CO_2. However, due to the distinct properties of H_2 gas compared to conventional CO gas, the reduction behavior varies depending on the gas composition. H_2 gas has a higher diffusivity than CO gas, which affects the reduction behavior. Furthermore, H_2 reduction is characterized by endothermic reaction heat, while CO reduction is associated with exothermic reaction heat. Therefore, it is necessary to analyze the different reduction behaviors of iron ore under various gas atmosphere conditions containing CO and H_2. Additionally, the analyses should be scaled up from the single particle to the moving packed bed, considering the distribution of gas atmosphere within the furnace. Therefore, in this study, a novel numerical analysis model has been proposed, developed, and validated to analyze the reduction behavior of iron ore particles, addressing the limitations of conventional particle reduction analysis models. Additionally, a packed bed analysis model has been proposed and investigated to analyze the large-scale moving bed.

The analysis model for the reduction of a single iron ore particle, as named 3D reduction model, has been proposed as shown in Fig. 1. This model is designed to address the analysis of irregularly shaped particles by considering 3Dimensional systems. By considering transient gas diffusion and heat transfer in an unsteady

state, the model is capable of analyzing various intra-particle distributions, such as gas concentration, iron oxides, iron, reduction degree, and temperature, in the reduction behavior of iron ore.
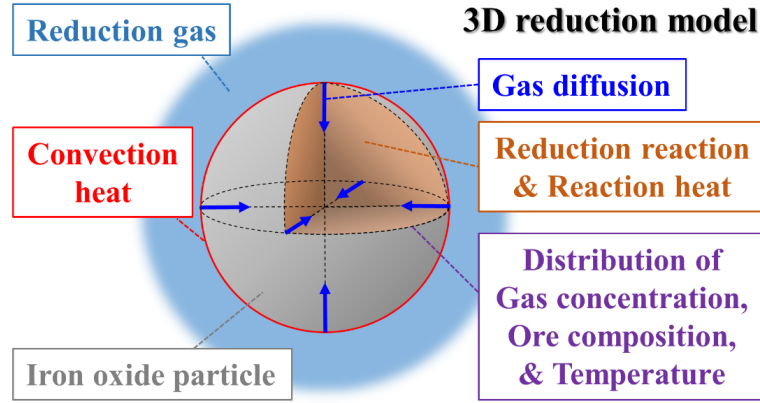

Fig. 1. Schematic of 3D-based single particle reduction model as named by 3D reduction model.

The governing equations utilized in the model include the 3D diffusion equation (Eq. 1) and heat transfer equation (Eq. 2), where the symbols represent C: Gas concentration [mol m^{-3}], C_P: Heat capacity [J kg^{-1} K^{-1}], D_e : Effective diffusivity [m^2 s^{-1}], Q' : Volumetric convection heat transfer rate [W m^{-3}], q_v : Volumetric reaction heat generation rate [W m^{-3}], R: Reaction rate [mol m^{-3} s^{-1}], t: Time [s], x, y, and z: Location in axes directions [m], i : Indirect reduction steps, j : Gas atmosphere, and λ_e: Effective thermal conductivity [W m^{-1} K^{-1}]. These equations respectively analyze the behavior of indirect reduction reactions and particle temperature considering by heat transfer of reaction heat and convection heat.

$$\frac{\partial C_j}{\partial t} = \frac{\partial}{\partial x}(D_{e,j} \frac{\partial C_j}{\partial x}) + \frac{\partial}{\partial y}(D_{e,j} \frac{\partial C_j}{\partial y}) + \frac{\partial}{\partial z}(D_{e,j} \frac{\partial C_j}{\partial z}) - \sum_i R_{ij} \quad \cdots (1)$$

$$\rho C_P \frac{\partial T}{\partial t} = \frac{\partial}{\partial x}(\lambda_e \frac{\partial T}{\partial x}) + \frac{\partial}{\partial y}(\lambda_e \frac{\partial T}{\partial y}) + \frac{\partial}{\partial z}(\lambda_e \frac{\partial T}{\partial z}) + q_v + Q' \quad \cdots (2)$$

The model results are compared and analyzed with experimental measurements conducted under reproduced experimental conditions. The model achieves an accuracy of over 98.3% when compared to the experimental measurements. Due to its high accuracy, the model can be utilized as a digital twin to assess reduction behaviors and derive reaction parameters.

  It is investigated the effect of particle shape on reduction behaviors by analyzing intra-particle distributions as shown in Fig. 2. The results of the analysis show that particles with a larger surface area exhibit an increased reduction degree over time, such as having a smaller sphericity, a higher number of macroscopic pores, and a jagged surface. This is attributed to the greater gas diffusion within the particle, as the concentration of diffused gas inside the particle drives the chemical reaction rate. Therefore, the reduction

behaviors seems to be similar to the gas diffusion behaviors. Specifically, this larger surface area, characterized by a jagged surface, results in an increased gas diffusion rate through 3Dimensional diffusion directions and subsequently faster reduction degrees over time.
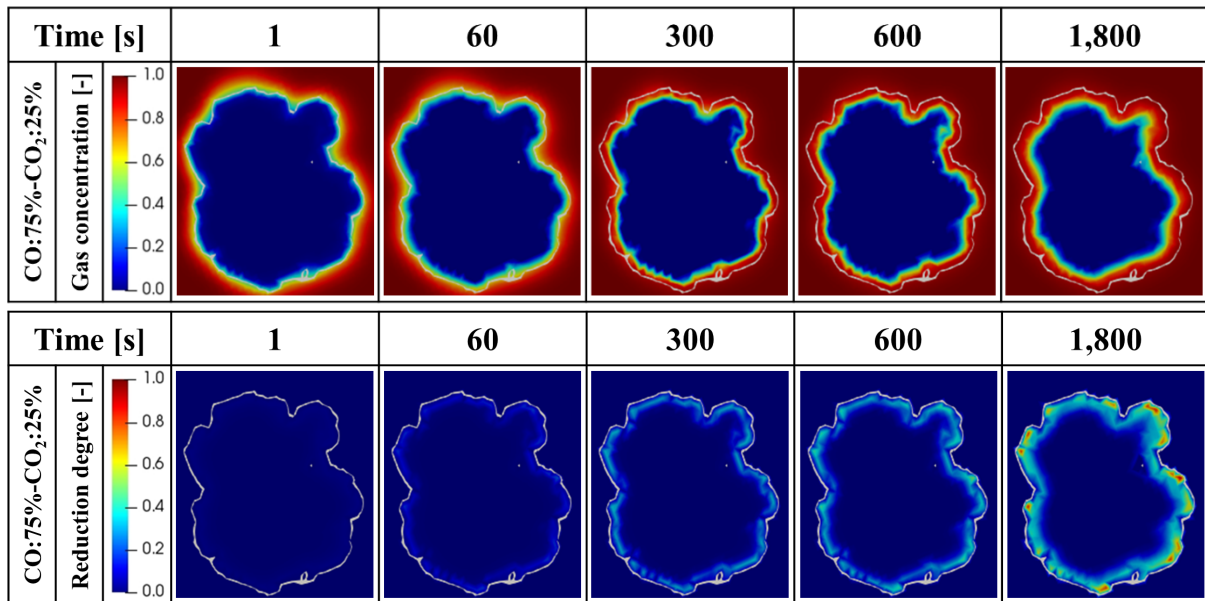

Fig. 2. Intra-particle distribution of gas concentration and reduction degree in sintered ore shaped particle.

It is discussed the impact of H_2 on the reduction behavior of iron ore particles and analyzes the intra-particle distribution as demonstrated in Fig. 3. It is observed a reaction front behavior is observed in CO reduction with reaction surface, while in the presence of H_2, the reaction behavior becomes broader and more widespread throughout the particle. It is found that the reduction degree over time increases when H_2 is present in the CO-H_2 atmosphere under the same driving force conditions, because the H_2 component has a significant impact on the effective gas diffusivity, which has a greater influence on the reduction rate compared to the chemical reaction rate constant. Therefore, the widespread reduction behaviors by a higher H_2 concentration result in an increase in the reduction degree over time.
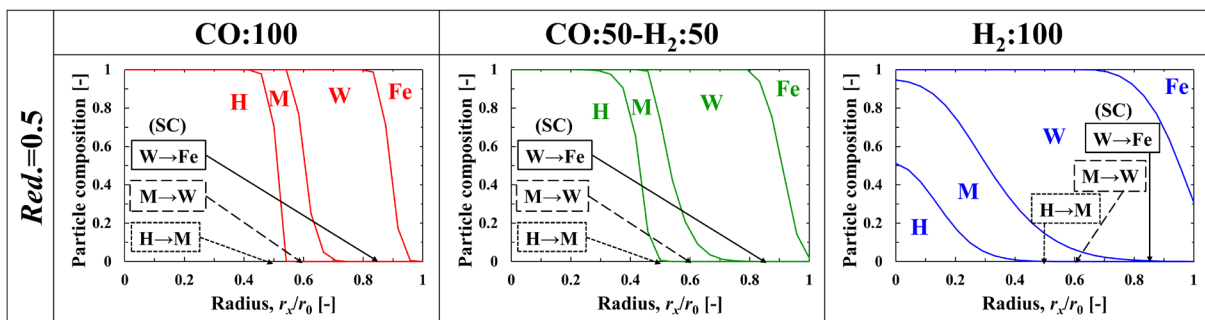

Fig. 3. Particle compositional distribution in CO, CO-H_2, and H_2 atmospheres in the 3D reduction model and the certain reaction surface in the conventional model.

It is analyzed the effect of particle temperature through heat transfer during reduction reactions under the blast furnace condition. When heat transfer is considered, including both reaction heat and convection heat, the particle temperature tends to converge to the temperature of the reduction gas over time, because the convective heat transfer rate becomes higher than the reaction heat generation rate over time. Due to the higher heat transfer coefficient in H_2 reduction compared to CO reduction, the effect of convective heat transfer becomes more prominent in H_2 reduction, while the effect of reaction heat becomes more significant in CO reduction. Increasing particle size causes a significant intra-particle temperature changes, primarily due to the effect of reaction heat. It is concluded that the effect of reaction heat on particle temperature change becomes prominent with the particle size.

It is necessary to scale up from the single particle to the moving packed bed in order to analyze the reduction behavior of iron ores. The packed bed exhibits gaseous distribution of temperature and composition, leading to different reduction behaviors of iron ore within the same iron ore layers. In the moving bed, the gaseous temperature leads to the solidus particle temperatures. In the coke layer, the temperature decreases compared near iron ore layers due to the endothermic of coke reactions. In the iron ore layer, the temperature follows the indirect reaction heat. The reduction behavior of the iron ore is influenced by the gas atmosphere. In particular, the H_2-based atmosphere exhibits a much higher reduction degree at the same time compared to the CO-based atmosphere, despite the iron ore layer temperature being higher in the CO-based atmosphere. These results indicate that the gas atmosphere plays a crucial role in driving the reduction rate.

To mitigate CO_2 emissions in industry, the introduction of the hydrogen ironmaking process is crucial. In this study, the distribution of iron ore reduction with respect to the H_2 reductant has been clarified through numerical analysis. The reductant gas composition impacts the reduction behavior due to a broader distribution of the H_2 component. This widespread reduction behavior results in a faster reduction rate, despite its endothermic heat. Moreover, the reduction rate and process temperature are dependent on the H_2 concentration in the reductant gas atmosphere. It is essential to optimize the H_2 concentration maintaining the optimal temperature for hydrogen ironmaking process.