Potentials and limitations of low-carbon steelmaking process: Iron ore reduction with hydrogen in shaft furnace

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Abstract

A promising future approach to reducing CO2 emissions in the metallurgical sector is to reduce iron oxides using renewable hydrogen in a shaft furnace. Here, the potentials and limitations of iron ore reduction with hydrogen at high and low reduction temperatures were studied through two-level dynamic modeling: (i) combined modeling of gas/solid reaction, transfer, and diffusion in porous iron ore pellet, (ii) modeling of the shaft furnace reactor on an industrial scale. A transient one-dimensional reactor model is developed and coupled to the pellet model. In order to predict the conversion and energy performance of the reactor, the conversion and energy efficiency were established on the scale of an industrial reactor for two contrasted operating conditions: (1) at 900 °C, which ensures the complete conversion of iron ore into pure iron with rapid conversion kinetics, (2) at 550 °C, which is more compatible with the use of renewable energy sources for heating.

**Keywords**: iron ore direct reduction, modeling of gas-solid reaction and diffusion, hydrogen, porous solids.

* 1. Introduction

Metallurgy is an emblematic energy-consumption process in our society. The reduction of iron oxide to metallic iron, steel, and cast-iron using coal is the process that gave rise to the industrial revolution, clearly taking advantage of the high energy density contained in cheap coal despite its considerable environmental and societal impacts. The production of 1.7 billion tons of steel per year (2017) results in the release of twice as much CO2 (7 % of global emissions) (*Iron & Steel*, s. d.). The concept of utilizing hydrogen as a reducing agent, as genuinely reviewed in (Spreitzer & Schenk, 2019), is primarily connected to the concern of climate change because the reduction of iron oxide with hydrogen produces water vapor rather than the carbon dioxide produced by the reduction with the carbon monoxide (coal) (Patisson & Mirgaux, 2020). Around 80 % of the world’s pre-reduced iron is produced using a gaseous mixture of hydrogen and carbon monoxide as the reducing gas in a shaft furnace. The first production of pre-reduced iron using pure hydrogen is scheduled for 2025. Therefore, our aim is to show the potentials and limitations of such processes using pure hydrogen at pellet and reactor scale when operating at different operating conditions, such as the reduction temperatures, iron ore pellet size, and water content in the reducing gas.

* 1. Methods

In the reactor, the various mass and energy transport phenomena are coupled with chemical reactions. In order to predict the process performance, a pellet-scale model is first developed to simulate the various transfer, diffusion, and reaction phenomena. Then, this model is integrated into the global reactor model that simulates the conversion and thermal behaviors at a large scale. The following pellet and reactor models consist of homemade Python scripts.

* + 1. Pellet model

The pellet model is based on the progression of a shrinking core by involving simultaneously reaction-diffusion phenomena at each reacting solid layer. This model is commonly applied to situations where a chemical reaction occurs at the surface of a solid material, and over time, the reaction progresses inward, causing the solid core to "shrink" as the reaction front moves deeper into the material. When the reactant transforms into another solid material, leaving behind an unreacted solid, the unreacted shrinking core model (USCM) is applied (Levenspiel, 1998). Since the reduction of iron oxides with hydrogen involves four reactions (R1-4) as a function of temperature, the USCM is extended to consider a wide range of reduction temperatures, from low temperatures (T < 570 °C) where wüstite (FeO) is unstable up to higher temperatures. Consequently, the kinetics of chemical reactions are established separately at low and high temperatures with their respective stoichiometries.

From a chemical reaction perspective, first-order reaction rates (Ri), as shown in Eq. (1), are formulated according to characteristic dimensions, i.e., the radius of the pellet (r0) and the radius of the corresponding solid layer (ri), the kinetic constant of reaction (ki), and the gas concentrations ($C\_{H\_{2}}$, $C\_{H\_{2}O}$). Note that reactions can be reversible depending on the chemical equilibrium constant ($K\_{i}$). In order to determine the conversion of iron ore pellets, the dynamic evolution of each solid layer is then determined in terms of solid concentration ($C\_{s,k}$) as a function of reaction rates and stochiometric coefficients ($ν\_{k,i}$) through Eq. (2).

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| $$Fe\_{2}O\_{3}+H\_{2}\rightarrow 2Fe\_{3}O\_{4}+H\_{2}O$$ | (R1) |
| $$Fe\_{3}O\_{4}+H\_{2}\rightarrow 3FeO+H\_{2}O$$ | (R2) |
| $$FeO+H\_{2}\rightarrow Fe+H\_{2}O$$ | (R3) |
| $$Fe\_{3}O\_{4}+4H\_{2}\rightarrow 3Fe+4H\_{2}O$$ | (R4) |

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| --- | --- |
| $$R\_{i}=3 \frac{r\_{i}^{2}}{r\_{0}^{3}} k\_{i}\left(C\_{H\_{2}}\left(r\_{i}\right)-\frac{C\_{H\_{2}O}\left(r\_{i}\right)}{K\_{i}} \right)$$ | (1) |

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| --- | --- |
| $$\frac{∂C\_{s,k}}{∂r}=\sum\_{i}^{reactions}ν\_{k,i}.R\_{i} $$ | (2) |

Nonetheless, calculating reaction rates requires knowledge of the pellet’s radial gas concentration ($C\_{j}$). For this purpose, the pseudo-steady-state approach for the gas diffusion is used for each solid layer, as shown in Eq. (3) assuming a spherical pellet:

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| $$\frac{1}{r^{2}}\frac{∂}{∂r}\left(r^{2}D\_{eff,j}\frac{∂C\_{j}}{∂r}\right)=0$$ | (3) |

Where r is the corresponding radius and Deff,j is the effective diffusivity of gas j in the solid layer resulting from Bosanquet’s approach (Krishna & Van Baten, 2012). At the pellet boundary condition (r = r0), the diffusion is equal to the amount of mass transfer between the pellet and the bulk gas, as formulated in Eq. (4), where the mass transfer coefficients ($k\_{m,j}$) are obtained using Sherwood correlation for spherical pellet (E, 1952).

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| --- | --- |
| $$D\_{eff,j}\left(\frac{∂C\_{j}}{∂r}\right)=k\_{m,j}\left(C\_{j,bulk}-C\_{j}\left(r\_{0}\right)\right)$$ | (4) |

* + 1. Reactor model

The reactor model describes the transport, chemical conversion, and thermal effects involved in a countercurrent vertical furnace where cold solid is supplied at the top while the hot gas is injected at the bottom. The model is based on solving the mass and energy conservation equations by including the pellet model (diffusion and reaction with temperature-dependent kinetics). The model is transient and one-dimensional with respect to the height of the reactor. The mass balance equations for all gas and solid components, as well as the energy balance equations for the gas and solid phases are formulated based on the general convection-diffusion equation derived from the general continuity equation as shown in Eq. (5), where the extensity $θ$ consists of the mass fraction for the mass balances and the specific enthalpy for the energy balances (Ranzani Da Costa et al., 2013).

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| --- | --- |
| $$\frac{∂}{∂t}\left(ρθ\right)+div\left(ρθ\vec{u}\right)-div\left(D.\vec{∇}θ\right)=S$$ | (5) |

Where $ρ$ is the density, D the dispersion coefficient, and u the velocity.

The first three terms in Eq. (5) describe the accumulation, the convection, and the diffusion, respectively. S in Eq. (5) is the source term related to the chemical reactions involved both in gas and solid phases. The assumptions used here are: (i) Plug flow is considered for both phases; (ii) The reactor is assumed to be adiabatic; (iii) Uniform pellet temperature, size, and shape at any axial position in the reactor; (iv) Constant bed porosity; (v): Two distinct stoichiometric, at high and low temperatures, coexist in the reactor depending on the temperature profile; (vi): Two types of heat transfer are considered: conduction and convection.

In addition, by assuming an average bed porosity ($ε$), and by combining the Ergun equation (Wagner, 2008) and the continuity equation in the gas phase, the axial profile of gas velocity (ug) and pressure were established in the bed.

* 1. Results and discussion

Although higher reduction temperature results in a higher conversion of iron ore to pure iron (Fig. 1 (a)), studying the apparent conversion kinetics for a single iron ore pellet reveals that an optimal conversion kinetics can be found at a lower temperature (550 °C) (Pescott, 1976). The latter is potentially compatible with alternative low-tech heating solutions using renewable flow energies. Therefore, here the influence of main operating parameters such as hot gas inlet temperature and water vapor content on the conversion and temperature profile was studied to establish the actual conversion and energy efficiency on the scale of an industrial reactor.

Figure 1. shows that based on a water vapor content of 2 % in the inlet gas, when the inlet gas temperature decreases from 900 °C to 550 °C, the conversion decreases from 100 % to 54 % while the energy consumption for heating is reduced by 47 %. In addition, an increase in water vapor content markedly affects the conversion even at high temperatures: no complete conversion for a temperature of 900 °C and 550 °C with a water vapor content of 10 % in the inlet gas.



**Figure 1.** (a): Reduction curves for different inlet gas temperatures and water vapor content. (b): Temperature profile in the reactor for different inlet gas temperatures.

Figure 1. (b) shows that with an inlet gas temperature of 900 °C, the temperature is uniform in the reactor within 3 meters of the gas inlet. The hot gas provides an energy input to compensate the energy required for endothermic reduction reactions with hydrogen. The temperature profile at 550 °C coincides with the shape of the reduction curve at this temperature. In fact, the kinetics are strongly affected at lower temperatures and also by a progressive formation of water vapor through chemical reactions that significantly limit the conversion.

In comparison with the iron oxide reduction performance of a conventional shaft furnace operating with a gaseous mixture consisting mainly of hydrogen and carbon monoxide (50% H2, 32% CO, 9% CH4, 4% H2O, 2% CO2) : (i) a complete conversion is obtained with a more compact reactor in the case where pure hydrogen is used, this fact due to the more efficient kinetics; (ii) zero CO2 emissions in a hydrogen reactor since there is no direct CO2 production; (iii) almost the same order of magnitude of energy consumption is observed (4% smaller energy consumption using pure hydrogen as the reducing agent), note that the reduction with pure hydrogen is endothermic while in conventional shaft furnace operating with syngas involves also exothermic combustion reactions.

* 1. Conclusions

The model-based studies provided in this work helped to understand the potentials and limitations of direct iron ore reduction with hydrogen at a real reactor scale. By comparing a reactor fueled with pure hydrogen at 550 °C with the same reactor fueled at 900 °C, the following main observations were made: (i) Although the thermal requirement for heating decreases by 47 % when it operates at 550 °C, conversion drops from 100 % to 54 % for 10 mm iron ore pellets, (ii) temperature profile and conversion are greatly affected at lower temperatures due to loss of conversion kinetics, (iii) the presence of water vapor in the gas becomes the main thermochemical barrier for the final conversion of magnetite to pure iron at lower operating temperatures.

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