Application of Carbon Recycling Iron-making System in a Shaft Furnace

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An active carbon-recycling energy system (ACRES) has been proposed to reduce emission carbon dioxide (CO2) emission from industrial energy processes. Application of a smart iron-making system based on ACRES (iACRES) in a shaft furnace is modeled numerically as a new low-carbon process. It was assumed that a proportion of the CO2 in the furnace gas was extracted via gas separation, and reduced into carbon monoxide (CO) by electrolysis, after which regenerated CO was mixed with a reduction gas and recycled continuously in the furnace. The use of a solid oxide electrolysis cell (SOEC) was assumed for the electrolysis process. A one-dimensional model for the shaft furnace was employed for feasibility evaluation of the carbon recycling process.

The mixing ratio of electrolysis gas, \( m \) [-], was defined as the flow amount of CO2 separated from the furnace gas for recycling (which was electrolyzed into CO/CO2 mixture) relative to total inlet reduction gas for the furnace. Electrolysis degree, \( ed \) [-], was defined as CO production yield by electrolysis of the separated CO2. The effects of \( m \) and \( ed \) on the reduction process in the furnace were evaluated. At \( ed > 70\% \), metallization degree of > 90% was maintained at \( m > 10\% \). The furnace system was envisaged as a pre-reduction process for iron-ore material. When 70% metallization was acceptable for the pre-reduction process, \( m \) of 14% was achievable even at \( ed \) of 40%. The value of \( m \) is equal to primary fuel saving. It is expected that the shaft furnace with iACRES would have potential as a low-carbon iron-making process.

KEY WORDS: carbon recycling; iACRES; shaft furnace; carbon dioxide; carbon monoxide.

1. Introduction

Reductions in the consumption fossil resources and the emission of carbon dioxide are keenly required for future sustainability. An active carbon-recycling energy system (ACRES) has been proposed to reduce carbon dioxide (CO2) emission from industrial energy processes.1) Iron-making processes that consume large amounts of fossil resources show substantial potential for such reductions, for which carbon recycling is seen as a key technology. The possibility of a smart iron-making process based on the active carbon recycling energy system (iACRES) has been discussed.2) In the iACRES process, CO2 in furnace gas exhausted from an iron-making furnace is separated and reduced into carbon materials such as carbon monoxide (CO) and carbon (C) by a reduction process driven by non-fossil energy sources. Regenerated CO and C are recycled in the furnace. CO2 electrolysis is a primary candidate for CO2 reduction because it is capable of clearly separating reduced CO and oxygen (O2) at cathode and anode sides, respectively. A solid oxide electrolysis cell (SOEC) has been studied for CO2 or H2O reductions at 600–900°C.3) CO2 electrolysis using a solid polyelectrolyte fuel cell has been demonstrated by input over potential electricity at room temperature.4) At higher temperature, Gibb’s free energy change of CO2 is reduced and therefore CO2 electrolysis becomes more efficient.5,6) As the primary energy for driving CO2 reduction in iACRES, heat output up to 950°C produced from a high-temperature gas-cooled reactor (HTGR) is the primary candidate, because HTGR output of around 600 MW is sufficiently large, and also stable and safe.7) The process of heat exchange between HTGR gas media and SOEC requires a temperature differential. It is thought that the output temperature of a HTGR is sufficient for SOEC operation at 900°C. A shaft furnace is expected to be more suitable for recycling CO generated by SOEC in iACRES than a blast furnace, because the shaft furnace consumes CO directly within the reduction process. The present study uses a one-dimensional numerical model of a shaft furnace to assess the feasibility of recycling CO in a shaft furnace with iACRES.
2. Analytical Model

2.1. Shaft Furnace with iACRES

A shaft furnace is used to assess the feasibility of iACRES, as shown in Fig. 1. Synthesized gas containing CO, hydrogen (H₂), water, nitrogen, and CO₂ mixture produced by methane steam reforming is supplied into the shaft furnace as a reduction gas. Iron ore is supplied into the furnace from its top, and direct-reduced iron (DRI) is produced by reaction with the synthesized gas. Hot briquette iron is finally made from the DRI. Pressure swing adsorption or amine absorption CO₂ separation processes are assumed for separation of CO₂ from the furnace gas. SOEC is a candidate for reduction by utilizing heat up to 950°C generated from HTGR. The resulting mixture of CO and CO₂ is combined with the inlet gas flow and recycled in the furnace. By-product O₂ is available for use in other oxidation processes.

2.2. Numerical Model

A one-dimensional numerical model of a shaft furnace was employed. Governing equations for the mass balances (Eqs. (1) and (2)) and energy balances (Eqs. (3) and (4)) of gas and solid phases are shown below:

\[
\frac{\partial}{\partial t} \left( \varepsilon_i \rho_i x_{ij} \right) + \frac{\partial}{\partial z} \left( \varepsilon_i \rho_i x_{ij} u_j \right) = \sum_a m_{a,ij} R_a \quad \text{(1)}
\]

\[
\frac{\partial}{\partial t} \left( (1-\varepsilon_i) \rho_i x_{ij} \right) + \frac{\partial}{\partial z} \left( (1-\varepsilon_i) \rho_i x_{ij} u_j \right) = \sum_a m_{a,ij} R_a
\]

\[
\frac{\partial}{\partial t} \left( \sum_{j} c_{ij} x_{ij} \right) + \frac{\partial}{\partial z} \left( \sum_{j} c_{ij} x_{ij} u_j \right) = \sum_{a} \rho_{a} \left( T_{a} - T \right) - \sum_{k} \gamma_k \frac{\partial}{\partial z} \left( \sum_{j} c_{kj} x_{kj} u_j \right) \quad \text{(2)}
\]

\[
\frac{\partial}{\partial t} \left( T(1-\varepsilon_i) \rho_i x_{ij} \right) + \frac{\partial}{\partial z} \left( T(1-\varepsilon_i) \rho_i x_{ij} u_j \right) = \sum_{a} \rho_{a} \Delta H_a \left( T_a - T \right) - \sum_{k} \gamma_k \frac{\partial}{\partial z} \left( \sum_{j} c_{kj} x_{kj} T_j \right) + h_a \frac{4}{D} \left( T_{w} - T \right) - \Delta H_e \sum_{k} m_{a,ik} R_a \quad \text{(3)}
\]

Here, \( \varepsilon_i \) [-], \( \rho_i \) [kg m⁻³], \( x_{ij} \) [-], \( u_j \) [m s⁻¹], \( m_{a,ij} \) [-], \( R_a \) [kmol m⁻³ s⁻¹], and \( C_{ij} \) [J g⁻¹ K⁻¹] present void fraction of furnace bed, material density, component composition, velocity, reaction stoichiometric coefficient, reaction rate, and component’s specific heat, respectively. The subscripts s, g, n indicate solid and gas phases, and reaction number, as shown in Table 1. \( j \) (j = 1–5) represent CO, CO₂, H₂, H₂O, N₂, respectively. \( sj \) (j = 1–7) represent FeO₂–, FeO₄, FeO, Fe, iron ore gangue, moisture contents, and C, respectively. \( h_b \) [J s⁻¹ m⁻² K⁻¹] and \( h_w \) [J s⁻¹ m⁻² K⁻¹] are the heat transfer coefficient between gas and solid, and overall heat transfer coefficient through the furnace wall, respectively. \( a \) [m⁻²] and \( D \) [m] are the specific surface area of raw material of iron ore and diameter of furnace, respectively. \( T \) [K] and \( \Delta H_e \) [kJ mol⁻¹] indicate temperature and reaction enthalpy. These equations were solved via the Euler–Gauss method. Table 1 shows chemical reaction rates (\( R_a \) [kmol min⁻¹ m⁻bed⁻¹]) considered in the one-dimensional shaft furnace model, and rate constants (\( k_i \) [m s⁻¹]) of these reactions for sintered ore. CO and H₂ reductions, carbon deposition, shift reaction, and water evaporation for iron ore pellet were considered. Reaction rate constants for Eqs. R₁ to R₆ in Table 1 were obtained by application of triple-phase shrinking core model to the kinetic results measured by fixed-bed type reactor experiments for sintered ore. Kinetic constants of carbon deposition of Eq. R₇ were decided from a fixed-bed experiment. Shift reaction of Eq. R₈ was assumed as reaction equilibrium. The validity of the numerical model was previously demonstrated via agreement between measured results of a fixed-bed type experiment and a shaft furnace test operation with calculation results based on the model. That reaction model is subsequently used in the present study. The operating conditions assumed for the furnace are shown in Table 2.

Figure 1 shows flow amounts of separated CO₂ from furnace gas for recycling: \( \dot{Q}_{e-CO₂} \) [Nm³ t-HM⁻¹]; CO produced from electrolysis of the separated CO₂: \( \dot{Q}_{e-CO₂} \) [Nm³ t-HM⁻¹]; CO remaining after electrolysis: \( \dot{Q}_{s-CO₂} \) [Nm³ t-HM⁻¹]; and

![Fig. 1. A shaft furnace with iACRES.](image-url)
total inlet reduction gas in the furnace: \( Q_{\text{b-total}} \) [Nm\(^3\) t-HM\(^{-1}\)].

Electrolysis degree, \( ed \) [%], was defined as CO production yield by electrolysis of the separated CO\(_2\) as in Eq. (5):

\[
ed \% = \frac{Q_{e-CO}}{Q_{e-CO} + Q_{e-CO_2}} \times 100 \tag{5}
\]

Electrolysis efficiency of water into hydrogen was reported as 82%.\(^{11}\) Due to a lack of previous studies on the efficiency of CO\(_2\) electrolysis into CO, it was assumed that \( m > 80\% \) was available for CO\(_2\) electrolysis by a SOEC as well as water electrolysis. The volumetric mixing ratio of electrolysis gas, \( m \) [%], was defined as the CO\(_2\) separated from furnace gas (reduced to electrolysis gas of CO and CO\(_2\) mixture) relative to total inlet reduction gas for the furnace.

\[
m \% = \frac{Q_{e-CO}}{Q_{b-total}} \times 100 \tag{6}
\]

that is,

\[
Q_{e-CO} = \frac{Q_{e-CO} + Q_{e-CO_2}}{Q_{b-total}} = (m/100)Q_{b-total} \tag{7}
\]

\( Q_{b-total} \) was fixed at 1 500 Nm\(^3\) t-HM\(^{-1}\) constantly in this calculation. Inlet reformed gas flow amount: \( Q_{\text{ref}} \) [Nm\(^3\) t-HM\(^{-1}\)] which had the reformed gas composition shown in Table 2 was decided from the following equation.

\[
Q_{\text{ref}} = \left[1 - \left(\frac{m}{100}\right)\right]Q_{b-total} = Q_{b-total} - Q_{e-CO_2} \tag{8}
\]

In Eqs. (6) and (8), \( m \) represents the reduction of inlet reformed gas ratio, and primary fossil consumption and consequent CO\(_2\) emission. Mass balances and energy balances in the shaft furnace were calculated using parameters of \( ed \) and \( m \) via the one-dimensional model. Figure 2 shows changes in the furnace inlet compositions of CO and CO\(_2\) according to changes in \( m \) and \( ed \). CO utilization factor (\( \eta_{CO} \)) and H\(_2\) utilization factor (\( \eta_{H_2} \)) at a section in the shaft furnace are defined as follows:

\[
\text{CO utilization factor} \% = \eta_{CO} = \frac{x_{CO}}{\left(x_{CO} + x_{CO_2}\right)} \times 100 \tag{9}
\]

\[
\text{H}_2 \text{ utilization factor} \% = \eta_{H_2} = \frac{x_{H_2}}{\left(x_{H_2} + x_{H_2O}\right)} \times 100 \tag{10}
\]

\( x_{g_j} \) shows component composition of the gas of \( g_j \) (\( j = 1-4 \)) at a local section in the furnace. The availability of CO recycling in the shaft furnace system was estimated by calculation of these values.

### Table 2. Shaft furnace conditions for numerical analysis.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Raw material charge rate [t-raw d(^{-1}) m(^{-3})]</td>
<td>12.7</td>
</tr>
<tr>
<td>Production rate [t-HM d(^{-1}) m(^{-3})]</td>
<td>7.3</td>
</tr>
<tr>
<td>Shaft furnace volume [m(^3)]</td>
<td>150</td>
</tr>
<tr>
<td>Effective height of packed bed in the furnace [m]</td>
<td>9.0</td>
</tr>
<tr>
<td>Reformed gas composition [volume %]</td>
<td>CO = 30, CO(_2) = 15, H(_2) = 45, H(_2)O = 0, N(_2) = 10</td>
</tr>
<tr>
<td>Total inlet reduction gas flow amount [Nm(^3) t-HM(^{-1})]</td>
<td>1 500</td>
</tr>
<tr>
<td>Inlet reduction gas temperature [°C]</td>
<td>900</td>
</tr>
<tr>
<td>Heat loss through furnace</td>
<td>0 (insulated)</td>
</tr>
<tr>
<td>Iron ore material</td>
<td>Sintered ore</td>
</tr>
</tbody>
</table>

(mean diameter 10.5 mm)

Fig. 2. Change of furnace inlet compositions of CO and CO\(_2\) by change of \( m \) and \( ed \). (Online version in color.)

3. Results and Discussion

The calculated reduction and metallization degree distributions, solid temperature, CO and H\(_2\) utilization factors, and corresponding reduction rates in a shaft furnace are shown in Fig. 3. The ordinate shows depth from the stock line. The lower and upper dashed lines show the positions of the tuyere and stock line, respectively. Figures 3(a-1) and 3(a-2) represent a conventional shaft furnace without furnace gas recycling (non-recycled case). Figures 3(b-1) and 3(b-2) represent the case with furnace gas recycling at \( ed = 60\% \) and \( m = 30\% \). Reduction degree for iron ore was defined as the oxygen removed during the reaction relative to the initial oxygen content of the ore. Metallization degree for iron ore was defined as the amount of metallic iron produced relative to the total iron content of the ore. Reduction and metallization degrees of the recycled case are inferior to those of the non-recycled case, because the inlet \( ed \) value of the recycled case is less than the inlet \( ed \) of 67% for the non-recycled case. Subsequently, in the recycled case, the
rates of CO and H₂ reduction at the tuyere at the bottom of the bed are smaller than those of the non-recycled case.

The effects of mixing and electrolysis degrees on gas utilization factors are shown in Fig. 4. Figure 4(a) shows the effect of mixing ratio, $m$, from 0 to 30% on $\eta_{\text{CO}}$ and $\eta_{\text{H}_2}$. At larger $m$, there is less change in $\eta_{\text{CO}}$ at the tuyere at the bottom of the bed, because CO₂ concentration at $m = 30\%$ is more similar to the equilibrium value than at lower $m$. Figure 4(b) shows the effect of electrolysis degree at lower $m$ on $\eta_{\text{CO}}$ and $\eta_{\text{H}_2}$. At higher $ed$, there is greater change in $\eta_{\text{CO}}$ at the tuyere because the inlet gas has higher CO concentration. $\eta_{\text{CO}}$ at $ed = 40\%$ declines near the tuyere, because a reverse-shift reaction from CO₂ and CO proceeds until reaching reaction equilibrium. Ultimately, both $\eta_{\text{CO}}$ and $\eta_{\text{H}_2}$ reach similar values at the top of the bed. CO and H₂ reduction rates [kmol s⁻¹ m-bed⁻³] were defined as total rate of $R_1 + R_2 + R_3$ and $R_4 + R_5 + R_6$, respectively. Figure 5 shows the effect of $ed$ on reduction rates for (a) CO, and (b) H₂. The scenario with larger $ed$ shows higher rates of CO and H₂ reaction because CO and H₂ inlet concentrations are higher at higher $ed$. Particularly at $ed = 80\%$, reduction rates are superior to the non-gas recycling case.

Figure 6 shows effect of $ed$ on (a) metallization degree
and (b) reduction degree at the tuyere line of the bed. At ed of 90%, metallization degree reaches 100% at more than 15% of m. This demonstrates an upper limit of m for carbon recycling. At ed of less than 70%, the metallization degree gradually decreases with increase of m due to declining CO content. Reduction degree in Fig. 6(b) shows a similar trend to Fig. 6(a), and demonstrates that the synthesized gas mixed with electrolysis gas is capable of reducing iron ore. Figure 7 shows the effects of ed on metallization degree and m. Pre-reduction of iron ore is useful in some iron-making processes such as COREX,[12] which aims to achieve m of around 90%. When metallization degree of 90% was available for a pre-reduction process, it was possible to realize the degree at m of more than 30% and ed of more than 80%, as shown in Fig. 6(a); and at m of 2.3, 3.1, 4.8, and 10.0%
with \(ed\) of 40, 50, 60, 70%, respectively, as shown in Fig. 7. When metallization degree of 70% is available for a pre-reduction process, it is possible to realize the degree at \(m\) of 14% at \(ed\) of 40%. The value of \(m\) is a direct indication of reduction of reformed gas of \(Q_{\text{ref}}\) and consumption of primary fossil material, and hence CO\(_2\) emission. It was confirmed that installation of an iACRES system in a shaft furnace had the capability to reduce consumption of primary fossil material, and to mitigate CO\(_2\) emission under the operating conditions examined.

4. Conclusions

The model results confirm that the installation of an iACRES system in a shaft furnace has the capability to reduce consumption of primary fossil material, and to mitigate CO\(_2\) emission. A furnace fitted with iACRES driven by primary energy from non-carbon sources such as HTGR is expected to be appropriate for pre-reduction processes. When metallization degree of 90% was required for the furnace, it was calculated that more than 30% reduction in primary carbon consumption and CO\(_2\) emission could be achieved at \(ed\) of more than 80%, and 10% was available at \(ed\) of 70%. When metallization degree of 70% was acceptable for the pre-reduction process, \(m\) of 14% was sufficient even at \(ed\) of 40%. The findings suggest a shaft furnace with iACRES as a promising candidate for low-carbon iron-making processes.

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REFERENCES