Process Evaluation of Use of High Temperature Gas-cooled Reactors to an Ironmaking System Based on Active Carbon Recycling Energy System

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Reducing coking coal consumption and CO₂ emissions by application of iACRES (ironmaking system based on active carbon recycling energy system) was investigated using process flow modeling to show effectiveness of HTGRs (high temperature gas-cooled reactors) adoption to iACRES. Two systems were evaluated: a SOEC (solid oxide electrolysis cell) system using CO₂ electrolysis and a RWGS (reverse water-gas shift reaction) system using RWGS reaction with H₂ produced by iodine-sulfur process. Both reduction of the coking coal consumption and CO₂ emissions were greater in the RWGS system than those in the SOEC system. It was the reason of the result that excess H₂ not consumed in the RWGS reaction was used as reducing agent in the blast furnace as well as CO. Heat balance in the HTGR, SOEC and RWGS modules were evaluated to clarify process components to be improved. Optimization of the SOEC temperature was desired to reduce Joule heat input for high efficiency operation of the SOEC system. Higher H₂ production thermal efficiency in the IS process for the RWGS system is effective for more efficient HTGR heat utilization. The SOEC system was able to utilize HTGR heat to reduce CO₂ emissions more efficiently by comparing CO₂ emissions reduction per unit heat of the HTGR.

KEY WORDS: carbon recycling iron and steel production; high temperature gas-cooled reactor; CO₂ electrolysis; reverse water gas shift reaction; process flow analysis; CO₂ emissions.

1. Introduction

Import price of coking coal in Japan, which is used as coke source in BF (blast furnace, abbreviations are summarized at the end of this paper, the same applies hereinafter) ironmaking, is increasing steeply in the last decade by significant increase of steel production in developing countries, China in particular. Ironmaking processes with less coking coal consumption are desired. Reduction of CO₂ emissions is now also demanded due to a concern of its impact on climate change. R&D is widely conducted aiming the emissions reduction from iron and steel production industry because this is one of the main CO₂ emissions sources.

Recycling BFG (blast furnace gas) to a BF, injection of by-product gases to a BF, CCS (carbon capture and storage) have been proposed and studied as methods to reduce coking coal consumption and CO₂ emissions. Application of ACRES (active carbon recycling energy system) to an iron and steel production process is also proposed for the purpose. ACRES is a concept of recycling carbon compounds of high exergy such as CO recovered from CO₂ emitted from industry.¹⁾ When exergy source of low CO₂ emissions is used for the recovery, fossil fuel consumption and CO₂ emissions are expected to be reduced by transfer of some part of exergy consumption of the fuels to the exergy of the

Fig. 1. Concept of iACRES.

source. The ironmaking process applying ACRES is called iACRES (ironmaking system based on ACRES).²⁾ Figure 1 shows the concept of iACRES. Fe₂O₃, the main compound in iron ore, is reduced to Fe as in reaction (1).

The CO₂ from reaction (1) is reduced to CO using exergy of heat, electricity and H₂. The recovered CO is used to reduce the Fe₂O₃ as in reaction (2).

Coke consumption in the BF is expected to be reduced by injection of the recovered CO into the BF. Consequently, coking coal input to a coke oven is reduced by the decrease

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of coke demand of the BF. CO₂ emissions can also be reduced as a result of the CO recovery from the BFG and decrease of by-product COG (coke oven gas) by the reduction of coke production.

A HTGR (high temperature gas-cooled reactor), a type of nuclear reactor featuring high temperature helium gas heat carrier and graphite moderators in the core, is a candidate of low CO₂ emissions exergy source. Advantages of HTGRs are as follows: high exergy density, high exergy efficiency with high reactor outlet temperature of 950°C, capacity to follow fluctuation of exergy demand.³⁾

CO₂ electrolysis (reaction (3)) and CO₂ reduction with H₂ in RWGS (reverse water gas shift) reaction (reaction (4)) are proposed as candidates of the CO recovery.⁴⁾

CO₂ electrolysis is advantageous in high temperature in thermodynamics as explained in section 2.2. SOEC (solid oxide electrolysis cells) are used to electrolyze CO₂ in high temperature over 800°C.⁵⁾ RWGS is an equilibrium reaction progressing at around 600°C without side reactions with Cu compound catalyst.⁶⁾ The temperatures of the reactions match the maximum temperature of 950°C of HTGR reactor outlet helium. H₂ used for RWGS reaction is desired to be produced with little CO₂ emissions from both material source and exergy source. Water electrolysis and thermochemical water splitting are considered as candidates of such H₂ production methods.

Heat and material balance in CO₂ electrolysis by SOEC^{2,4,5,7-9)} and CO₂ reduction by RWGS reaction⁴⁾ were investigated as for iACRES. Iron ore was assumed to be reduced by only the recovered CO in the most of the studies.^{2,4,5,7,9)} The supposition was different from actual operation condition of BFs using coke and PC (pulverized coal). In addition, these studies assumed that all of CO₂ in BFG was reduced to CO. This meant no CO₂ was emitted from the process. In actual ironmaking processes, some part of BFG is consumed for heat supply to the process and electricity generation. Further, reducing all of the CO₂ in the BFG is difficult considering reaction equilibrium. Thus, effect of iACRES seems to be overestimated in these studies. Though Nakagaki

et al.,⁸⁾ considered coke and PC as reducing materials in addition to CO, neither coking coal consumption nor CO₂ emissions were evaluated since the investigated range of the study was only HTGRs and SOEC. Therefore, effect of iACRES on coal consumption and CO₂ emissions is not clear.

This study analyzed influence of iACRES on reduction of coking coal consumption and CO₂ emissions through process flow investigation of total BF iron and steel production systems in detail. The calculation outcome was compared with a one of a conventional BF iron and steel production process. Number of HTGRs requirement per BF and CO₂ emissions reduction per unit HTGR heat input were evaluated to show applicability of the HTGR to iACRES. CO₂ electrolysis in SOEC and CO₂ reduction by RWGS were considered as CO recovery methods. Thermochemical water splitting IS (iodine-sulfur) process was adopted as H₂ production.

2. Process Flow Simulation of iACRES Systems

Process flow simulation of iACRES systems was done to investigate their heat, electricity and material balance. A commercial chemical process simulator Aspen Plus (Aspen Technology, Inc.) ver. 7.3 was used for the analysis. A system adopted CO₂ electrolysis (SOEC system) and a system using IS process H₂ production and RWGS CO₂ reduction (RWGS system) were evaluated and outcomes were compared with outcomes of a conventional BF system. **Figure 2** illustrates the schematic diagrams of the SOEC and the RWGS systems. Total systems are composed of modules of HTGR, SOEC, IS process, RWGS and BF. Heat and material balance of the total systems were calculated by fitting heat and material inputs and outputs between modules. Heat and material balance were normalized to produce HM (hot metal) of 1 ton from the BF.

Heat and material balance per BF was proportional to the ones of the 1 ton HM case. HM production rate per BF was determined as 1.22×10^{-1} t-HM/s (identical to 3.83×10^3 kt-HM/y) assuming a large scale BF as for Japan of inner volume of 5 000 m³ and relatively high productivity of 2.1 t-HM/(d·m³).¹⁰⁾

Coking coal input and CO₂ emissions were evaluated to show effects of iACRES using HTGRs. Heat and electricity demands in the SOEC, IS process and RWGS modules were

Fig. 2. Schematic diagram of the SOEC system and the RWGS system.

calculated to show number of HTGRs requirement for one BF. CO₂ emissions per unit HTGR heat in the SOEC and the RWGS systems were compared.

2.1. HTGR Module

HTGRs supplied exergy for CO₂ reduction directly as heat and electricity and indirectly as H₂ produced in the IS process.

A HTGR process flow model for the SOEC system was constructed using design conditions of GTHTR300C (gas turbine high temperature reactor 300C), a type of HTGR which provides heat and electricity at the same time.³⁾ **Figure 3** shows the model of the HTGR module. The primary helium gas was heated in the reactor to 1223.15 K. High temperature heat of the helium was transferred first to the secondary helium loop through the IHX (intermediate heat exchanger) and then to CO₂ in the SOEC and in the pre-heater. Heat of the primary helium at the outlet of the IHX was used to generate electricity by the helium gas turbine. The electricity was consumed in the SOEC and CO₂ separation. Excess electricity was transmitted outside of the iron and steel production system. **Table 1** summarizes operation conditions of the module.

No process flow model of a HTGR module was made for the RWGS system. HTGR heat (Q_{Reactor}) was calculated from heat ($Q_{\text{IS,RWGS}}$) and electricity ($W_{\text{IS,RWGS}}$) consumed in IS process and RWGS modules assuming electricity generation thermal efficiency defined in Eq. (5) as 46.8%.¹²⁾

$$\eta_{el.} = \frac{W_{\text{IS,RWGS}}}{Q_{\text{Reactor}} - Q_{\text{IS,RWGS}}} \times 100 \quad (5).$$

2.2. SOEC Module

SOEC module recovered CO by electrolysis of CO₂ separated from BFG and recycled the CO into a BF.

Figure 4(a) shows the simplified process flow model of the SOEC module. This module is mainly composed of CO₂ separation columns and SOEC. Some part of the BFG from the BF was consumed in combustion to supply heat to blast in heat stoves and used in power generation to supply electricity to the BF module. Here, 30% of the BFG was split to CO₂ separation for recycling. CO₂ of 90% was separated

from the split BFG using chemical absorption method.¹¹⁾ The BFG contacted solvent in an absorption column to make CO₂ solved into the solvent. The solvent was heated in a stripping column reboiler to vaporize the CO₂ and recover the solvent. The off gas from the stripping column was consumed just as the BFG to supply process heat and electricity. The CO₂ from the CO₂ stripping column was reduced to CO in the SOEC. Conversion ratio of CO₂ in the SOEC was fixed at 60% to match the one in RWGS, whose conversion ratio was limited by reaction equilibrium. Table 1 shows operation conditions.

Temperature requirement for the CO₂ stripping column reboiler was 413.15 K. If heat of the HTGR secondary helium loop of 1223.15 K had been transferred to the reboiler, exergy loss would have been large because temperature difference between the second helium and the reboiler was large. Instead, waste heat from the BF module was supplied to the reboiler.

SOEC temperature (T) was determined as 1073.15 K considering factors below. Electricity corresponding to reaction Gibbs energy (ΔG) and heat corresponding to reaction heat ($T\Delta S$) should be supplied to progress the CO₂ electrolysis reaction. Relation of the values is shown in Eq. (6),

$$\Delta H = \Delta G + T\Delta S \quad (6).$$

The value of ΔG is smaller at higher T because reaction enthalpy (ΔH) and reaction entropy (ΔS) depend little on the T and the value of ΔS is plus. Even though high T is advantageous for small electricity input, the T had upper limit due to the reactor temperature of the HTGR. Heat of the HTGR reactor at 1223.15 K was transferred to the SOEC through two heat exchangers: the IHX and the SOEC itself. Therefore, T was set at 1073.15 K. Heat and electricity input to the SOEC were calculated as follows. SOEC operation voltage (E) was considered as sum of theoretical voltage (E_t) and over potential (E_o) as in Eq. (7),

$$E = E_t + E_o \quad (7).$$

E_t was determined by only T (0.981 V at 800°C) while E_o was influenced by not only the T but current density (i). E_o was calculated using current density dependence of E in an

Fig. 3. Flow model of the HTGR module in the SOEC system.

Table 1. Operation conditions of the HTGR module and the SOEC module in the SOEC system.

Module	Component	Parameter	Symbol	Unit	Note	Reference
HTGR	Primary He loop	Reactor heat per HTGR		MW _t	600	3)
		Reactor outlet temperature		K	1 223.15	3)
		IHX outlet temperature		K	1 204.82	d)
		Minimum temperature difference in the IHX		K	50	3)
	Secondary He loop	He temperature at the inlet of the SOEC		K	1 173.15	3)
SOEC	Before CO ₂ separation	BFG flow rate before splitting		kmol/s	8.5	d)
		BFG splitting ratio to CO ₂ separation ((Stream S-3)/(Stream S-1)) in Fig. 4(a)		mol%	30	e)
	CO ₂ separation columns	CO ₂ recovery ratio ((Stream S-5)/(Stream S-3)) in Fig. 4(a)		mol%	90	a)
		Recovery ratio except CO ₂		mol%	0	e)
		Temperature of heat input to the stripping column reboiler		K	413.15	b)
	SOEC	CO ₂ conversion ratio		mol%	60	e)
		Current	I	A	7.45E+07	d)
		Current density	i	A/cm ²	0.24	d)
		Theoretical voltage	E_t	V	0.98	c)
		Over potential	E_o	V	0.34	d)
		Operation voltage ($E = E_t + E_o$)	E	V	1.32	d)
		Cell temperature	T	K	1 073.15	e)

 a) Typical value of a conventional MEA (monoethanolamine) absorption process¹¹⁾

 b) Above 20 K of a typical temperature of a conventional MEA absorption process (393.15 K) in Ref. ¹¹⁾

c) Determined from only temperature

d) Result of this study

e) Given specifications

Fig. 4. Simplified flow model of the SOEC, RWGS and BF modules.

experiment at 800°C as in Eq. (8),¹³⁾

$$E = 1.58 \times i^{0.126} \dots\dots\dots (8).$$

The part of electricity consumption corresponding to the theoretical voltage ($E_d I$) was equal to the ΔG . On the other hand, the part corresponding to the over potential ($E_o I$) was consumed as Joule heat. Heat input from the HTGR was calculated by subtracting the Joule heat from the reaction heat ($T\Delta S - E_o I$). In actual calculation, flow rate of BFG inlet and i at the maximum value of the ($T\Delta S - E_o I$) were determined by process flow simulation. When the BFG inlet flow rate was set at a certain value, ΔS and I were determined because reaction amount of CO_2 was set with fixed conversion ratio in the SOEC at 60%. Next, when the value of i was set at a value, E was defined by Eq. (8) and E_o was determined by Eq. (7). The heat input from the HTGR ($T\Delta S - E_o I$) was calculated using the values determined above. BFG inlet flow rate and i were varied to find the maximum value of the HTGR heat input.

2.3. IS Process Module

H₂ was produced from water using heat and electricity from the HTGR in the IS process module. The H₂ was fed to the RWGS reaction to reduce CO₂. IS process is comprised of the three chemical reactions below:

H₂O is decomposed into H₂ and O₂ in the total process while the other compounds cycle in the process.

The highest temperature requirement was 1 173.15 K in Eq. (10); temperatures of the other heat inputs were lower than that. Sensible heat of the second helium loop in the HTGR was expected to be utilized in a stepwise fashion.

Figure 5 shows the simplified flow model of the IS process. SO_2 from the H_2SO_4 section and I_2 from the HI section reacted with H_2O to produce H_2SO_4 and HI. Product solution from the Bunsen reaction was separated automatically into $\text{H}_2\text{SO}_4\text{--H}_2\text{O}$ solution and HIx ($\text{HI--I}_2\text{--H}_2\text{O}$) solution.

The $\text{H}_2\text{SO}_4\text{--H}_2\text{O}$ solution was fed to the H_2SO_4 section. First, H_2SO_4 was concentrated in the H_2SO_4 concentrator by H_2O vaporization. In the H_2SO_4 recovery column, the concentrated H_2SO_4 contacted returning stream from the SO_2 decomposer and remaining H_2SO_4 in the stream was captured. H_2SO_4 in the bottom stream of the column was vaporized and decomposed into SO_3 and H_2O in the H_2SO_4 vaporizer. The SO_3 vapor was decomposed into SO_2 and O_2 in the SO_3 decomposer. The product returned to the H_2SO_4 recovery column. The SO_2 and O_2 gas returned to the Bunsen section from the top of the column. The SO_2 was reacted in the Bunsen reactor and the O_2 was obtained as by-product of the IS process. The HIx solution from the Bunsen section was first fed to the electro-electrodialysis cell, where HI was concentrated. The HI -rich solution from the cell flowed to the HI distillation column, where almost pure HI vapor was distilled from the top. The HI vapor was decomposed into H_2 and I_2 in the HI decomposer. The product I_2 and remaining HI were separated from the product gas in the I_2 separator and the HI separator, respectively. Small amount of impurities in the remaining gas was captured into water fed from outside. The H_2O was also used as reactant in the Bunsen reaction. Product H_2 was obtained as a result. All heat and electricity requirement were supplied from the HTGR. **Table 2** shows main design conditions of the module.

2.4. RWGS Module

In the RWGS module, CO₂ separated from BFG reacts with H₂ from the IS process to recover CO as in Eq. (4). This module contains CO₂ separation columns, a RWGS reactor and a H₂O condenser.

Figure 4(b) shows the simple diagram of the module. Just as in the SOEC module, 30% of BFG from the BF was split to CO₂ separation, and 90% of CO₂ was separated from the split BFG. Off gas from the CO₂ separation columns was consumed as BFG not fed to CO₂ separation just as in the case of the SOEC module. The separated CO₂ flowed to the RWGS reactor with H₂ from the IS process to produce CO and by-product H₂O. RWGS temperature was fixed at 913.15 K. Feed ratio of the H₂ to the CO feed was determined at 2.55 to make CO₂ conversion ratio 60% in the reaction equilibrium at the temperature. Some part of the by-product H₂O was condensed in the H₂O condenser to be removed before injection into the BF because the H₂O have negative effects on BF heat balance. The other compounds

Fig. 5. Simplified flow model of the in process module.

Table 2. Operation conditions of the IS process module in the RWGS system.

Section	Component	Parameter	Symbol	Unit	Note	Reference
Bunsen	Bunsen reactor	Temperature		K	373.15	b)
H ₂ SO ₄	Inlet	Flow rate (H ₂ SO ₄ : H ₂ O)		mol/s	1 : 5.14	c)
	H ₂ SO ₄ vaporizer	Temperature		K	800.15	b)
	SO ₃ decomposer	Temperature		K	1 120.15	b)
		SO ₃ conversion ratio		mole fraction	0.615	a) c)
HI	Inlet	Flow rate (HI : I ₂ : H ₂ O)		mol/s	13.2 : 78.7 : 60.9	c)
	Electro-electrodialysis cell	Flow rate to the HI distillation column (HI : I ₂ : H ₂ O)		mol/s	4.1 : 0.2 : 14.7	c)
	HI distillation column	Distillate rate (HI)		mol/s	8.7	c)
		HI fraction in distillate		mole fraction	0.999	b)
		Bottom temperature		K	420.03	c)
	HI decomposer	Temperature		K	773.15	b)
		HI conversion ratio		mole fraction	0.230	a) c)
	H ₂ O feed	Flow rate (H ₂ O)		mol/s	1	b)
	H ₂ separator	H ₂ production		mol/s	1	b)
		Electricity generation thermal efficiency	$\eta_{el.}$	%	46.8	12)

a) Reaction equilibrium value calculated by Aspen Plus ver.7.3

b) Given specifications

c) Result of this study

Table 3. Operation conditions of the RWGS module in the RWGS system.

Module	Component	Parameter	Symbol	Unit	Note	Reference
	Before CO ₂ separation	BFG flow rate before splitting		kmol/s	8.3	d)
		BFG splitting ratio to CO ₂ separation ((Stream R-3)/(Stream R-1)) in Fig. 4(b)		mol%	30	c)
		CO ₂ recovery ratio ((Stream R-5)/(Stream R-3)) in Fig. 4(b)		mol%	90	a) 11)
RWGS	CO ₂ separation	Recovery ratio except CO ₂		mol%	0	c)
		Temperature of heat input to the stripping column reboiler		K	413.15	b)
		H ₂ /CO ₂ input ratio		Molar ratio	2.55	d)
	RWGS reactor	CO ₂ conversion ratio		mol%	60	c)
		Temperature		K	913.15	c)
		Electricity generation thermal efficiency	$\eta_{el.}$	%	46.8	12)

 a) Typical value of a conventional MEA absorption process¹¹⁾

 b) Above 20 K of a typical temperature of a conventional MEA absorption process (120°C) in Ref. ¹¹⁾.

c) Given specifications

d) Result of this study

(CO, CO₂ and H₂) and remaining H₂O were injected into the BF. **Table 3** summarizes operation parameters.

2.5. BF Module

Iron ore and sinter are reduced to produce steel in the BF module. This study set the range of investigation as coking in coke ovens, sintering of iron ore fine in sintering machines, heat stoves for heating blast, BOF (basic oxygen furnace) steelmaking, casting and rolling in addition to BF as shown in Fig. 2. A process flow model of the BF and the heat stoves was made based on operation data of a BF steel-making plant. Operation data of the other process components in the plant was applied to the evaluation. Figure 4(c) shows the simplified diagram of the modeled part.

Table 4 shows main operation conditions. Operation

conditions of the BF were different among systems due to different amount of reducing materials made by material recycling in the iACRES systems. PC input amount to the BF was fixed in any systems. On the other hand, coke input amount was changed by the different operation conditions. Coke was considered as pure carbon; PC was assumed as mixture of C, H₂, H₂O and metal oxides. Some part of by-product gases of COG, BFG and LDG (Linz-Donawitz converter Gas) was combusted to supply heat to the BF module. All the remaining gas was consumed in a gas-turbine¹⁴⁾ to generate electricity. Electricity requirement in the module was supplied sufficiently by the electricity and the excess one was transmitted outside.

Detail of the process flow model and simulation are discussed in a reference.¹⁰⁾

Table 4. Operation conditions of the BF module.

Component	Parameter	Unit	BF system	SOEC system	RWGS system	Note	Reference
Coke oven	Coal input	kg-coal/t-coke	1.61E+03	1.61E+03	1.61E+03		
BF	Coke input	kg-coke/t-HM	376	359	337		
	Pulverized coal input	kg-PC/t-HM	101	101	101		
	CO input	kg-CO/t-HM	–	89	82		c)
	H ₂ input	kg-H ₂ /t-HM	–	0	19		c)
	HM production	kt-HM/y	3.83E+03	3.83E+03	3.83E+03	a)	
	BFG production	kg-BFG/t-HM	2.21E+03	2.19E+03	1.95E+03		c)
By-product power generation	Electricity generation thermal efficiency	%	48.7	48.7	48.7	b)	14)

a) Determined by relatively high productivity (2.1 t-HM/(d·m³)) of standard large scale BF's (inner volume 5 000 m³) in Japan in Ref. ¹⁰⁾.

b) Higher heating value

c) Result of this study

Table 5. Material balance in the conventional BF.

		Unit: kmol/t-HM									
Stream No. in Fig. 4(c)		CO ₂	CO	H ₂ O	H ₂	CH ₄	N ₂	O ₂	(C)	(H)	
B-1	BFG	16.4	14.8	1.9	3.0	0.1	36.8				
B-2	Coke								27.9		
B-3	PC			0.1					7.1	5.6	
B-4	Air, O ₂ , Moisture			1.5			36.8	10.9			

(C) and (H) denote carbon and hydrogen elements that can be used for iron ore reducing.

Metal oxides in PC are not shown.

Table 6. Material balance in the SOEC module and the BF.

		Unit: kmol/t-HM									
Stream No. in Fig. 4(a)		CO ₂	CO	H ₂ O	H ₂	CH ₄	N ₂	O ₂	(C)	(H)	
S-1	BFG	19.6	15.7	2.0	2.9	0.1	30.1				
S-2	BFG	13.7	11.0	1.4	2.0	0.0	21.1				
S-3	BFG	5.9	4.7	0.6	0.9	0.0	9.0				
S-4	Off gas	0.6	4.7	0.6	0.9	0.0	9.0				
S-5	CO ₂	5.3									
S-6	O ₂							1.6			
S-7	SOEC outlet	2.1	3.2								
S-8	Coke								26.7		
S-9	PC			0.1					7.1	5.6	
S-10	Air, O ₂ , Moisture			1.5			30.1	10.9			

(C) and (H) denote carbon and hydrogen elements that can be used for iron ore reducing.

Metal oxides in PC are not shown.

3. Results and Discussion

3.1. Material Balance in SOEC/RWGS Modules and Coking Coal Input to the Iron and Steel Production Processes

Material balance except Fe and impurity metal elements in the BF, SOEC and RWGS modules is summarized.

Table 5 shows material balance in the conventional BF system. Coke of 27.9 kmol/t-HM was input to the BF system (stream B-2 in Fig. 4(c)). **Table 6** exhibits mass bal-

Table 7. Material balance in the RWGS module and the BF.

		Unit: kmol/t-HM									
Stream No. in Fig. 4(b)		CO ₂	CO	H ₂ O	H ₂	CH ₄	N ₂	O ₂	(C)	(H)	
R-1	BFG	18.1	14.7	4.8	8.0	0.5	22.4				
R-2	BFG	12.6	10.3	3.4	5.6	0.4	15.7				
R-3	BFG	5.4	4.4	1.5	2.4	0.2	6.7				
R-4	Off gas	0.5	4.4	1.5	2.4	0.2	6.7				
R-5	CO ₂	4.9									
R-6	H ₂				12.4						
R-7	CO ₂ , H ₂	4.9			12.4						
R-8	RWGS outlet	2.0	2.9	2.9	9.5						
R-9	H ₂ O			2.1							
R-10	BF inlet	2.0	2.9	0.8	9.5						
R-11	Coke								25.1		
R-12	PC			0.1					7.1	5.6	
R-13	Air, O ₂ , Moisture			0.7			22.4	10.9			

(C) and (H) denote carbon and hydrogen elements that can be used for iron ore reducing.

Metal oxides in PC are not shown.

ance in the SOEC module and BF in the SOEC system. CO of 3.2 kmol/t-HM was recycled into the BF (stream S-7 in Fig. 4(a)). Coke input to the BF was reduced to 26.7 kmol/t-HM from that of the conventional BF system because the recycled CO replaced some part of the coke. **Table 7** displays material flow rates in the RWGS module and BF in the RWGS system. Outlet stream of the RWGS reactor (stream R-8 in Fig. 4(b)) contained CO of 2.9 kmol/t-HM as a RWGS product and H₂ of 9.5 kmol/t-HM as remaining compound. The reducing materials in the recycled stream decreased coke input to 25.1 kmol/t-HM.

Table 8 shows coke input to the BF and coking coal input to the BF module to produce the coke. Recycling of reducing materials in the SOEC system and the RWGS system decreased the coke input from that of the conventional BF system. Coking coal input to the iron and steel production system was decreased from 541 kg/t-HM in the conventional BF system. The reductions were 23 kg/t-HM in the SOEC system and 56 kg/t-HM in the RWGS system. The reduction ratios of the coke input were 4.3% in the SOEC system and 10.3% in the RWGS system. Coking coal input reduction

Table 8. Coke input to the BF's and coking coal input to the BF modules.

	Unit	BF system	SOEC system	RWGS system
Coking coal input to coke oven	kg-coal/t-HM	541	518	486
Coking coal input reduction	kg-coal/t-HM	–	23	55
Coking coal input reduction ratio	%	–	4.3	10.3
Coke input to BF	kg-coke/t-HM	335	321	301
Coke input reduction	kg-coke/t-HM	–	14	34
Coke input reduction ratio	%	–	4.3	10.3

Table 9. CO₂ emissions from the iron and steel production systems.

	Unit	BF system	SOEC system	RWGS system	Note
CO ₂ from BFG, CO ₂ separation off gas	kmol/t-HM	31.2	30.0	28.4	
CO ₂ from the other sources	kmol/t-HM	12.5	12.2	11.8	a)
Total CO ₂ emission	kmol/t-HM	43.7	42.3	40.2	a)
Total CO ₂ emission	kg/t-HM	1.92E+03	1.86E+03	1.77E+03	a)
CO ₂ emissions reduction	kg/t-HM	–	65	158	a)
CO ₂ emissions reduction ratio	%	–	3.4	8.2	a)

a) C element in steel and chemicals from coke oven are not included.

ratio was the same as the one of coke input because the coking coal input amount was proportional to the coke input.

Total amount of the reducing materials, CO and H₂, recovered in the SOEC and RWGS modules were 3.2 kmol/t-HM and 12.4 kmol/t-HM, respectively. Though the total reducing materials in the RWGS system was about 3.9 times of those in the SOEC system, reduction of the coke input was 2.4 times. Material balance of the iron source feed, coke, PC, air, O₂, moisture and the recycled materials into the BF and the product BFG suggested contribution of C element to iron source reduction was larger than that of the same mole of H element. This was why the ratio of the coke input reduction in the RWGS system to that in the SOEC system was smaller than the ratio of the recycled reducing materials, CO and H₂, between these systems. It is effective to reduce coking coal input to avoid feeding H element to the BF. If the H₂ in the RWGS outlet stream is separated and recycled to the RWGS, the H₂ originally fed to the BF can be converted to CO₂ reduction. Consequently, less H₂ requirement for the IS process is expected and more efficient use of HTGR heat is anticipated. Membrane separation and pressure swing absorption methods are considered as candidates of H₂ separation.¹⁵⁾

3.2. CO₂ Emissions

Table 9 shows the effect of introduction of the SOEC and RWGS on CO₂ emissions reduction. The CO₂ emissions were mainly from COG by-product in coke production and BFG made in the BF. In the conventional BF system, all the BFG from the top of the BF was consumed. All C element in the BFG was eventually emitted as CO₂ (stream B-1 in Fig. 4(c)); the emissions were 31.2 kmol/t-HM. The other CO₂ emissions including the one from COG were 12.5 kmol/t-HM. Total CO₂ emissions from the module were 43.7 kmol/t-HM (1.92×10³ kg/t-HM). In the SOEC system and RWGS system, C element in the BFG split at the top of the BF and off gas from CO₂ separation were emitted from the systems as CO₂. All the C compounds in the split BFG (stream S-2 in Fig. 4(a)) and the CO₂ separation off gas (stream S-4) emitted as CO₂ of 30.0 kmol/t-HM in the SOEC system. CO₂ emissions from the other sources were 12.2 kmol/t-HM. Total CO₂ emissions were 42.3 kmol/t-HM (1.86×10³ kg/t-HM). Similarly, all of the C compounds in

the split BFG (stream R-2 in Fig. 4(b)) and the off gas from CO₂ separation (stream R-4) were eventually emitted as CO₂ of 28.4 kmol/t-HM in the RWGS system. CO₂ of 11.8 kmol/t-HM was emitted from the other part of the BF module besides the BF. CO₂ emissions from the system in total were 40.2 kmol/t-HM (1.77×10³ kg/t-HM). The emissions from the BFG were reduced as recycling some part of CO₂ in the BFG in the SOEC and RWGS systems. At the same time, CO₂ emissions from COG were reduced due to the decrease of coke consumption in the BF. CO recovery newly added in the SOEC and RWGS systems did not lead to the CO₂ emissions increase because the HTGR used for the CO recovery emitted no CO₂. As a result, total CO₂ emissions from the SOEC and RWGS systems were reduced from the conventional BF system.

CO₂ emissions reduction from the conventional BF system was 65 kg/t-HM in the SOEC system and 158 kg/t-HM in the RWGS system as in Table 9. Reduction ratio was 3.4% in the SOEC system and 8.2% in the RWGS system. The emissions reduction in the RWGS system was 2.4 times of that in the SOEC system. The ratio was similar to the ratio of coking coal input reduction because COG, BFG and CO₂ separation off gas, which occupied most of the CO₂ emissions, were originally from coking coal.

Considering the BFG splitting ratio to CO₂ separation, the CO₂ recovery ratio in the CO₂ separation, and the CO₂ conversion ratio in the SOEC and RWGS, only 16% of the CO₂ in the BFG was recycled as CO. This small percentage made the CO₂ emission reduction ratios relatively small. Higher BFG splitting ratio in the range of possible BF operation is required for more CO₂ emissions reduction since improvement of the reaction ratio is difficult and the CO₂ recovery ratio is already near 100%.

3.3. Heat and Electricity Balance

Heat balance in the HTGR, SOEC and RWGS modules were evaluated to clarify process components to be improved. HTGRs requirement per BF was also studied in these systems.

Table 10 shows heat and electricity balance of the HTGR module and the SOEC module in the SOEC system. Total heat and electricity demands of the SOEC module were 137 MJ/t-HM and 833 MJ_e/t-HM, respectively. The heat input

Table 10. Heat and electricity balance of the HTGR module and the SOEC module in the SOEC system.

			Heat	Electricity	Heat	Electricity	Note
			MJ _t /t-HM	MJ _e /t-HM	MW _t per BF ^{a)}	MW _e per BF ^{a)}	
Demand	SOEC	CO ₂ separation	930	24	113	3	b)
		Pre-heating of reducing gas	51		6		
		SOEC	86	809	10	98	
		Total demand	137	833	17	101	c)
		Total heat input considering electricity generation	1.87E+03		227		c)
		Excess electricity		287		35	d)
	Total demand for HTGR		137	1.12E+03	17	136	
Total heat requirement including excess electricity generation		2.47E+03		300			
			Heat	Electricity	Heat	Electricity	Note
			MJ _t /t-HM	MJ _e /t-HM	MW _t per BF ^{a)}	MW _e per BF ^{a)}	
Supply	HTGR	Heat supply	137		17		
		Electricity generation		1.12E+03		136	
		Reactor heat	2.46E+03		299		e)

a) HM of 1.22×10^{-1} t-HM/s is produced per BF.¹⁰⁾

b) Heat for CO₂ separation is supplied from waste heat in the BF module.

c) Heat for CO₂ separation and excess electricity are not included.

d) Transmitted outside

e) The value is not identical to sum of the heat input and the heat for electricity generation ("Total heat requirement including excess electricity generation" column) because of round-off error.

did not include heat to the CO₂ stripping column reboiler of 930 MJ_t/t-HM because sensible heat of exhaust and slag in the BF module was sufficient to supply the heat demand of the reboiler. Total HTGR heat consumption was 1.87×10^3 MJ_t/t-HM when the electricity input was converted to heat for electricity generation. Electricity generation for the SOEC occupied 93% of the HTGR heat: 1.73×10^3 MJ_t/t-HM of heat for electricity was consumed. **Table 11** shows a breakdown of the heat and electricity input to the SOEC. The heat supplied from the HTGR was only 86 MJ_t/t-HM, which was equivalent to the sensible heat of primary helium loop in the temperature range between reactor outlet (1 223.15 K) and IHX outlet (1 204.82 K). The heat was not enough for reaction heat ($T\Delta S$) of 295 MJ_t/t-HM. Electricity for Joule heat (E_oI) of 209 MJ_e/t-HM should also be supplied from electricity generated in the gas turbine of the HTGR. The Joule heat was not negligible compared with electricity for reaction (ΔG) of 600 MJ_e/t-HM. The Joule heat was desired to be less because it made an exergy loss in electricity generation step. Heat requirement of a SOEC ($T\Delta S$) is small at lower SOEC temperature as shown in Eq. (6), and HTGR can provide larger amount of heat ($T\Delta S - E_oI$) at lower SOEC temperature because wider temperature range of the primary helium between reactor outlet and IHX outlet is available. Consequently, Joule heat requirement (E_oI) is expected to be smaller. On the other hand, electricity requirement (ΔG) is larger at lower SOEC temperature as shown in Eq. (8). Determination of optimum temperature of SOEC (T) considering these factors is a problem for high efficiency operation.

Table 12 summarizes heat and electricity input from the HTGR module to the IS process and the RWGS modules in the RWGS system. Total heat and electricity input to the modules were 8.09×10^3 MJ_t/t-HM and 816 MJ_e/t-HM, respectively. Heat input to CO₂ separation was not counted just as in the case of SOEC system; the heat was recovered from sensible heat of exhaust and slag in the BF module. Total HTGR heat consumption was 9.84×10^3 MJ_t/t-HM

Table 11. Breakdown of heat and electricity balance in the SOECs.

		Heat	Electricity	Heat	Electricity
		MJ _t /t-HM	MJ _e /t-HM	MW _t per BF ^{a)}	MW _e per BF ^{a)}
Demand	ΔG		600		73
	$T\Delta S$	295		36	
		Heat	Electricity	Heat	Electricity
		MJ _t /t-HM	MJ _e /t-HM	MW _t per BF ^{a)}	MW _e per BF ^{a)}
Supply	Electricity input for ΔG		600		73
	Electricity for Joule heat (E_oI)		209		25
	HTGR heat input ($T\Delta S - E_oI$)	86		10	
Total		86	809	10	98

a) HM of 1.22×10^{-1} t-HM/s is produced per BF.¹⁰⁾

when the electricity input was converted to heat for electricity generation. Heat of 7.95×10^3 MJ_t/t-HM and electricity of 793 MJ_e/t-HM were used to produce H₂ in the IS process module. When the electricity input was converted to heat for electricity generation, HTGR heat for the H₂ production was 9.64×10^3 MJ_t/t-HM. High thermal efficiency was desired for H₂ production because the IS process module occupied 98% of the HTGR heat consumption.

Number of HTGRs required for a BF was evaluated from the heat balance in Tables 10 and 12. The HTGR heat including heat for electricity generation corresponded to 227 MW_t for a BF in the SOEC system. The number of HTGRs per BF was made to round ones with a little excess electricity generation transmitted outside. The BF required 0.5 HTGRs equivalent to heat of 300 MW_t. Electricity of 35

Table 12. Heat and electricity balance of the HTGR, IS process and RWGS modules in the RWGS system.

			Heat	Electricity	Heat	Electricity	Note
			MJ _t /t-HM	MJ _e /t-HM	MW _t per BF ^{a)}	MW _e per BF ^{a)}	
Module	Component						
Demand	IS process	H ₂ SO ₄ vaporizer	2.52E+03		306		
		SO ₃ decomposer	2.88E+03		350		
		HI distillation column	2.21E+03		269		
		HI decomposer	336		41		
		Electro-electrodialysis cell		625		76	
		Chiller in HI separator		73		9	
		Pumps		95		12	
	Total demand		7.95E+03	793	966	96	
	Total heat input considering electricity generation		9.64E+03		1.17E+03		
	RWGS	CO ₂ separation	858	22	104	3	b)
Pre-heating of reducing gas		43		5			
RWGS reaction		105		13			
Total demand		147	22	18	3	c)	
Total heat input considering electricity generation		195		24		c)	
Total demand of IS process and RWGS modules			8.09E+03	816	983	99	c)
Total heat input considering electricity generation			9.84E+03		1.20E+03		c)
Excess electricity				17		2	d)
Total demand for HTGR			8.09E+03	833	983	101	
Total heat requirement including excess electricity generation			9.87E+03		1.20E+03		
			Heat	Electricity	Heat	Electricity	Note
			MJ _t /t-HM	MJ _e /t-HM	MW _t per BF ^{a)}	MW _e per BF ^{a)}	
Module	Component						
Supply	HTGR	Reactor	9.87E+03		1.20E+03		

a) HM of 1.22×10^{-1} t/s is produced per BF.¹⁰⁾b) Heat for CO₂ separation is supplied from the BF module.c) Heat for CO₂ separation is not included.

d) Transmitted outside

Table 13. CO₂ emissions reduction per HTGR heat input.

	Unit	BF system	SOEC system	RWGS system	(RWGS system)/ (SOEC system)	Note
Total CO ₂ emissions	kg/t-HM	1.92E+03	1.86E+03	1.77E+03	—	
CO ₂ emissions reduction from the conventional BF system	kg/t-HM	—	65	158	2.4	
HTGR heat input	MJ _t /t-HM	—	2.46E+03	9.87E+03	4.0	a)
CO ₂ emissions reduction per HTGR heat input	kg/MJ _t	—	2.64E-02	1.60E-02	0.6	

a) Including excess electricity generation

MW_e per BF not used in the SOEC module was transmitted outside. A SOEC system iron and steel production plant composed of a HTGR and 2 BF_s was supposed. The HTGR heat taking the heat for electricity generation into consideration was 1.17×10^3 MW_t per BF in the RWGS system. The BF demanded 2 HTGRs equivalent to heat of 1.20×10^3 MW_t. Excess electricity of 2 MW_e per HTGR was transmitted outside. In the case, a unit RWGS iron and steel production plant was supposed to comprise 2 HTGRs and a BF.

3.4. CO₂ Emissions Reduction per Unit HTGR Heat Input

Table 13 summarizes CO₂ emissions per unit HTGR heat input. The CO₂ emissions reduction in the RWGS system from the conventional BF system (158 kg/t-HM) was 2.4

times of that in the SOEC system (65 kg/t-HM). The HTGR heat input to the RWGS system was 9.87×10^3 MJ_t/t-HM and that to the SOEC system was 2.46×10^3 MJ_t/t-HM including excess electricity generation. The heat input to the RWGS system was 4.0 times of that to the SOEC system. Consequently, the CO₂ emissions reduction per unit HTGR heat input of the SOEC system and the RWGS system were 2.64×10^{-2} kg/MJ_t and 1.60×10^{-2} kg/MJ_t, respectively. The SOEC system was able to utilize HTGR heat to reduce CO₂ emission more efficiently: the ratio of the reduction in the RWGS system to the SOEC system was 0.6. The larger HTGR heat input to the RWGS system than the SOEC system per unit HM production was the main reason of the smaller CO₂ emission reduction per unit heat in the RWGS system.

More efficient CO₂ reduction is required for the RWGS

system. H_2 production in the IS process per unit HM is desired to be reduced by recycling H_2 which was originally injected to the BF to the RWGS. If the recycling is achieved, HTGR heat for H_2 production per unit HM is reduced. Consequently, less CO_2 emissions per unit HM will be realized in RWGS system.

4. Conclusions

Reduction of coking coal consumption and CO_2 emissions by application of iACRES using HTGR exergy source to iron and steel production process was investigated by process flow modeling to clarify the effectiveness of the HTGR. Heat and material balance of a SOEC system and a RWGS system were calculated and the outcomes were compared with ones of a conventional BF system. Mass flow ratio of feed to CO_2 separation column to BFG was 30% and 90% of the CO_2 in the split BFG was separated in the columns. The CO_2 was electrolyzed to CO at the ratio of 60% in the SOEC system. On the other hand, the CO_2 was reduced in RWGS reaction to CO by H_2 produced in the IS process at the ratio of 60% in the RWGS system.

Coking coal consumption was reduced by 4.3% in the SOEC system and 10.3% in the RWGS system from the conventional BF system by substitution of coke input to a BF to the recovered CO. As the shift from coke to CO, CO_2 emissions were reduced by 3.4% in the SOEC system and 8.2% in the RWGS system. The following two factors were the reason of the emission reduction. Some part of the CO_2 in the BFG was recycled as CO recovered using exergy provided from the CO_2 -free HTGR; COG production was decreased as coke consumption in the BF was decreased. The reduction of the CO_2 emissions showed effectiveness of application of HTGRs to iACRES. The differences of the coking coal input reduction and CO_2 emissions reduction between systems were made due to the difference of returned reducing materials to the BF; only CO in the SOEC system, CO and H_2 in the RWGS system. Recycling the H_2 in the RWGS outlet stream to the RWGS is desired in the RWGS system because H element had smaller contribution to iron source reduction in the BF than C element did. Considering operation parameters, only 16% of CO_2 in the BFG was recycled as CO. Higher splitting ratio of BFG in the range of possible BF operation is required to achieve more CO_2 emissions reduction.

Heat balance in the HTGR, SOEC, IS process and RWGS modules were evaluated to clarify process components to be improved. HTGR heat of 2.46×10^3 MJ/t-HM was used in the SOEC module in the SOEC system, while HTGR heat of 9.87×10^3 MJ/t-HM was required for the IS process and RWGS modules in the RWGS system considering heat for electricity generation for the modules and excess electricity transmitted outside. Electricity generation for the SOEC occupied most of the HTGR heat. Some part of the electricity was consumed as Joule heat. Most of the HTGR heat usage was for H_2 production in the IS process in the RWGS system. Optimization of SOEC temperature in the SOEC system and high H_2 production thermal efficiency in the IS process in the RWGS system were desired for efficient HTGR heat utilization.

CO_2 emissions reduction per unit HTGR heat was 2.64×10^{-2} kg/MJ_t in the SOEC system and 1.60×10^{-2} kg/MJ_t in the RWGS system. The SOEC system was able to utilize HTGR heat to reduce CO_2 emissions more efficiently. It is desirable that recycling the H_2 injected to the BF to RWGS to reduce CO_2 to decrease H_2 requirement for the IS process and decrease CO_2 emission per unit HTGR heat.

Nomenclature

- E : Operation voltage of the SOEC (V)
- E_0 : Over potential in the SOEC (V)
- E_t : Theoretical voltage in the SOEC (= 0.981 (V))
- i : Current density in the SOEC (A/cm²)
- I : Current in the SOEC (A)
- $Q_{IS,RWGS}$: Heat input to IS process and RWGS modules (MJ/t-HM)
- $Q_{Reactor}$: Heat supply from nuclear reactor in HTGRs (MJ/t-HM)
- T : Temperature in the SOEC (K)
- $W_{IS,RWGS}$: Electricity input to IS process and RWGS modules (MJ/t-HM)
- ΔG : Reaction Gibbs energy of CO_2 electrolysis (MJ)
- ΔH : Reaction enthalpy of CO_2 electrolysis (MJ)
- ΔS : Reaction entropy of CO_2 electrolysis (MJ/K)
- η_{el} : Electricity generation thermal efficiency (%)

Subscripts

- _e: electricity
- _t: heat

Abbreviations

- ACRES: active carbon recycling energy system
- BF: blast furnace
- BFG: blast furnace gas
- BOF: basic oxygen furnace
- CCS: carbon capture and storage
- COG: coke oven gas
- GTHT300C: gas turbine high temperature reactor 300C
- HIx: mixture of HI–I₂–H₂O
- HM: hot metal
- HTGR: high temperature gas-cooled reactor
- iACRES: ironmaking system based on active carbon recycling energy system
- IHx: intermediate heat exchanger
- IS: iodine-sulfur
- LDG: Linz-Donawitz converter Gas
- MEA: monoethanolamine
- PC: pulverized coal
- RWGS: reverse water-gas shift reaction
- SOEC: solid oxide electrolysis cell

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