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THERMODYNAMIC ANALYSIS OF A NOVEL DIRECT REDUCTION IRON-H2 POLYGENERATION SYSTEM WITH CO2 CAPTURE

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ABSTRACT

In this paper, a novel system integrating direction reduction iron (DRI) production, H2 production, and CO2 capture has been proposed. The novel polygeneration system avoids excessive CH4 conversion rate, and the chemical energy of CH4 is cascade converted. Besides, the system realizes gradual enrichment of CO2 in the CH_4 conversion process. This paper presents the exergy analysis of the novel system. It shows that the energy saving ratio of the polygeneration system could reach up to 12.3%, and the performance improvements are mainly attributed to three aspects: cascade utilization of chemical energy, cascade utilization of physical energy, and lower separation penalty. For the polygeneration system, the CO2 concentration could reach up to 65%, and the energy consumption for separation is only about 30% of the general post-combustion separation process. At this point, the CO2 recovery ratio of the system is about 70%.

Keywords: DRI, CO2 capture, system integration

NOMENCLATURE

Abbreviations

CRR	CO_2 Recovery Ratio
DRI	Direction Reduction Iron
ESR	Exergy Saving Ratio
EUD	Energy Utilization Diagram
PSA	Pressure Swing Adsorption
WGS	Water Gas Shift

Symbols

E	Exergy
M	Mass Flow
n	Mole Flow
W	Power
γ	CH_4 consumption equivalent ratio of DRI production to H_2 production
η	Exergy Efficiency

1. INTRODUCTION

The steel industry is an important carbon emission source in China, and the technology of energy-saving and low-carbon has become the top priority of the steel industry[1]. The Direct Reduction Iron (DRI) process is regarded as the most promising technology. Its energy consumption and CO2 emission intensity are much lower than the ironmaking process in a blast furnace. Nowadays, the Midrex method is the leading process of DRI production technologies, and it has accounted for more than 60% of the DRI market[2]. For the Midrex DRI production process, the conversion rate of reducing gas and the carbon behavior during chemical reaction process are the most important factors affecting energy utilization efficiency and CO2 enrichment. Some researchers have done relevant researches in these two aspects. Liu et al. [3] deduced the formula for calculating the amount of reducing gas. Yi et al. [4] experimentally analyzed the influences of different reaction conditions such as reducing gas composition, reaction temperature and ore grade on the conversion rates. On the other hand, some researches[5] analyzed the carbon behaviors such as carbon deposition and carburization in the direct reduction process. On the basis, some shaft furnace simulation models were also gradually established[6], which provided necessary conditions for further exploration of the DRI process.

However, there are obvious irrational energy utilization in the DRI process, and the advantages of CO2 concentration in the DRI process cannot be fully exerted. Based on the principle of "cascade conversion of material according to composition", this paper proposes a DRI-H2 polygeneration system with CO2 capture, and analyzes the energy utilization of the polygeneration system.

2. PROPOSAL OF THE NOVEL SYSTEM

2.1 Basic concept of system integration

For the single DRI production process, the main goal is to increase the conversion rate of raw materials and maximize DRI production. In order to improve the CH4 conversion rate and DRI production, a large amount of unreacted gas should be recycled. When the CH4 conversion rate exceeds a certain value, the energy consumption for DRI will increase sharply. Therefore, exhaustion of the active composition causes great energy consumption for DRI production.

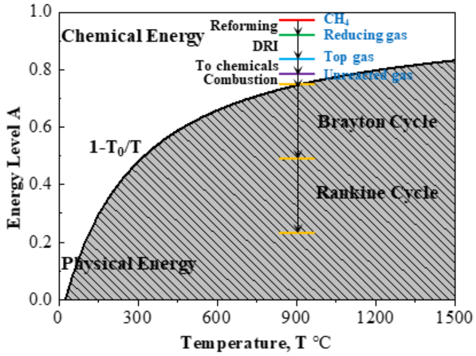


Fig 1 The principle of system integration

To overcome the problem of unreasonable energy utilization in the single DRI production system, the concept of system integration is proposed based on the principle of "cascade conversion of material according to composition", as shown in Fig 1. The CH4 with high energy level is first converted to reducing gas, and produces DRI and chemicals according to its component in cascade. And then, the unreacted gas is sent to generate heat by combustion or to generate power by combined cycle. By such cascade utilization of CH4, energy systems with high efficiency would be expected.

2.2 System integration

A conceptual flowsheet of the proposed polygeneration system is shown in Fig 2. CH4 is first reformed to reducing gas at 900℃ and 5 bar. Then the hot reducing gas is sent to the shaft furnace, whereafter DRI and top gas are output. After heat recovery, partial of the top gas is recycled to the reformer, and the rest part enters the water gas shift (WGS) reactors to concentrate H2 and CO2. Then the raw H2 is compressed to 25 bar and purified through pressure swing adsorption (PSA) process. Finally, most of the CO2 will be removed through the Selexol process. The rest gas mixed with CH4, CO and H2 will be used as the fuel of the external combustion unit. In the polygeneration system, DRI and H2 production are adjusted by varying the percentage of the recycled gas. Compared with the single DRI and H2 production process, the novel polygeneration is aimed at a higher energy utilization efficiency instead of the higher conversion rate of raw materials. Partial recycle of the unreacted gas is adopted in the polygeneration system, which avoids the excessive CH4 conversion and the sharp increase of energy consumption. For the polygeneration system, the chemical energy of CH4 is cascade converted, and the chemical energy level is gradually reduced with the output of different products. At the same time, the system realizes the gradual enrichment of CO2, and the CO2 could be captured with a low energy penalty.

3. EVALUATION CRITERIA

To evaluate the performance of the polygeneration system, the Midrex single DRI production system and the traditional single H2 production system are selected as the reference systems[7,8]. For the above systems, an external heat source should be provided. However, this paper pays more attention to the energy utilization of the CH4 conversion process. Consequently, the external heating process is not considered, and the parts surrounded by the red dotted line are selected as the research objects. The evaluation criteria and parameters are defined as follows.

3.1 Exergy efficiency

This paper uses the exergy analysis to evaluate the energy utilization of metallurgical-chemical processes. The exergy efficiency is defined below.

$$\eta = \frac{E_{H_2} + E_{DRI} + E_{Steam}}{E_{CH_4} + E_Q + W - E_{Fuel}} \tag{1}$$

Where E represents exergy, and the subscripts represent the corresponding substances. E_Q represents heat exergy, and W represents power.

3.2 CH4 consumption equivalent ratio of DRI production to H2 production

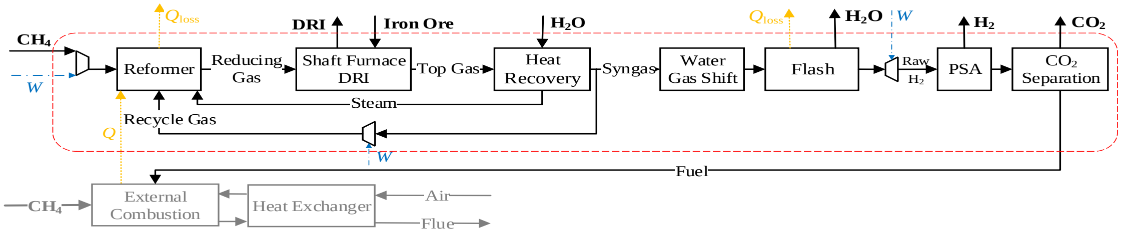


Fig 2 The flow diagrams of the DRI-H2 polygeneration system with CO2 capture

It is known that the amount of CH4 consumption for producing per mol DRI is 3/8 mol, and for producing per molar H2 is 1/4 mol. Therefore, the parameter γ is defined to denote the CH4 consumption equivalent ratio of DRI production to H2 production.

$$\gamma = \frac{{}^{3}\!/_{8}\, n_{\mathrm{Fe}}}{{}^{1}\!/_{4}\, n_{\mathrm{H_2}}} = \frac{3}{2}\frac{n_{\mathrm{Fe}}}{n_{\mathrm{H_2}}} \qquad (2)$$

Where n_{Fe} represents the molar production of Fe in DRI, and $n_{\mathrm{H_2}}$ represents the molar production of H2.

3.3 Exergy saving ratio

To compare the performance of the polygeneration system with single generation systems, exergy saving ratio (ESR) is defined as Eq.(3), where M_{H2} and M_{DRI} represent the mass flow of H_2 and DRI products; M_{CH4} and M_{Fuel} represent the mass flow of CH_4 input and Fuel output; EC_{DRI} and EC_{H2} denote exergy consumption for per mass products in the single DRI and H_2 production systems.

$$ESR = \frac{\left(M_{\mathrm{DRI}}\cdot EC_{\mathrm{DRI}} + M_{\mathrm{H_2}}\cdot EC_{\mathrm{H_2}}\right) - \left(M_{\mathrm{CH_4}}\cdot E_{\mathrm{CH_4}} - M_{\mathrm{Fuel}}\cdot E_{\mathrm{Fuel}}\right)}{M_{\mathrm{DRI}}\cdot EC_{\mathrm{DRI}} + M_{\mathrm{H_2}}\cdot EC_{\mathrm{H_2}}} \qquad (3)$$

3.4 CO2 recovery ratio

To evaluate the capability for CO2 recovery of the novel polygeneration system, CO2 recovery ratio (CRR) is defined by Eq.(4), which represents the percentage of carbon recovered to carbon input.

$$CRR = \frac{n_{\mathrm{CO_2}}}{n_{\mathrm{CH_4}}} \qquad (4)$$

Where $n_{\mathrm{CO_2}}$ and $n_{\mathrm{CH_4}}$ represent the mole flow of CO2 captured and CH4 input in the polygeneration system.

4. RESULTS AND DISCUSSION

4.1 System performance at the design condition

When the "Once-Through" strategy is employed, namely all the top gas enters the H2 producing side without recycling, the system performance is shown in Table 1. It is shown that the exergy efficiency of the polygeneration system is 85.1% and the ESR is 9.4%. Besides, the CRR of the polygeneration system reaches up to 70.4%.

Table 1. Performance of the DRI-H2 polygeneration

	Polygeneration	Single DRI Production	Single H2 Production
Exergy Input			
E_{CH4}, MW	830.5	278.3	533.2
Heat exergy, MW	196.6	154.9	173.3
Power, MW	66.6	51.4	16.4
Exergy Destruction			
In Total, MW	134.6	136.7	107.4
Exergy Output			
E_{H2}, MW	551.8	-	551.8
E_{DRI}, MW	215.3	215.3	-
E_{Fuel}, MW	192.0	122.6	63.6
E_{Steam}, MW	0.0	10.0	0.0
System Performance			
γ	0.33	-	-
η	85.1%	62.2%	83.7%
ESR	9.4%	-	-
CRR	70.4%	0.0%	0.0%

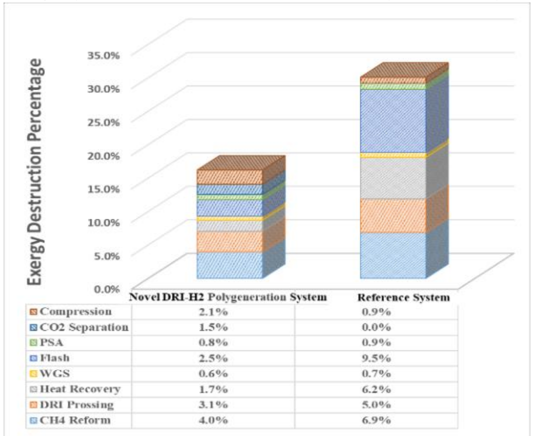
4.2 Exergy analysis at the design conditions

The energy saving mechanism of the new system could be disclose by using the exergy analysis method. In order to compare the exergy destruction of the novel polygeneration system and the single production systems in a more straightforward approach, the exergy destruction of each unit for the reference systems is combined, as shown in Fig 3. The comparison shows the energy saving performance of the polygeneration system is mainly reflected in three aspects.

The first aspect is cascade utilization of chemical energy. The polygeneration system series CH_4 reforming, shaft furnace DRI, and WGS units, and there is no need to pursue excessive raw material conversion rates. Eventually, the exergy destruction is reduced by 4.9 percentage points due to the cascade utilization of chemical energy.

The second aspect is cascade utilization of physical energy. The exergy destruction is reduced by 4.5 percentage points due to the reasonable utilization of physical energy.

The third part is derived from the separation processes. The separation process mainly includes Flash (phase separation), PSA (H2 component purification), Selexol (CO2 separation) and the related compression processes. The exergy destruction caused by Flash in the polygeneration system is much smaller. In addition, the CO2 concentration could be enriched to 65.0%, and the energy consumption for separation is only about 30% of the general post-combustion separation process. Eventually, the exergy destruction is reduced by 4.4 percentage points due to the lower separation penalty.



	Novel DRI-H2 Polygeneration System	Reference System
▨ Compression	2.1%	0.9%
▨ CO2 Separation	1.5%	0.0%
▨ PSA	0.8%	0.9%
▨ Flash	2.5%	9.5%
▨ WGS	0.6%	0.7%
▨ Heat Recovery	1.7%	6.2%
▨ DRI Prossing	3.1%	5.0%
▨ CH4 Reform	4.0%	6.9%

Fig 3 The distribution of exergy destruction for the polygeneration system and the reference systems

4.3 Sensitivity analysis

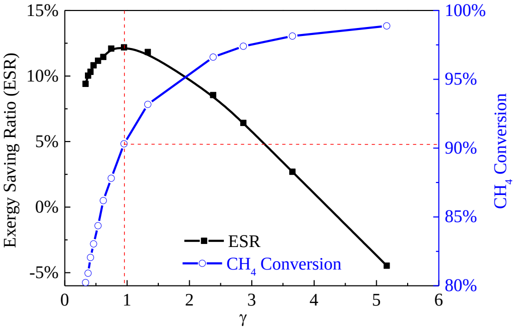


Fig 4 The effects of γ on ESR and CH4 conversion

Fig 4 is the effects of γ on ESR and CH4 conversion. As can be seen, γ varies from approximately 0.3 to 5.2 as the amount of recycle gas changes. At first, increasing γ leads to the increasing of ESR, because the energy saving potential of DRI production is higher than that of H2 production. But further increasing γ will cause high CH4 conversion ratio and thus the exergy destruction for DRI production increases sharply. Consequently, there is an optimized γ leading to the best system performance. When γ is about 0.95, the ESR of the polygeneration system could reach up to 12.3%, and the CH4 conversion rate is about 91%. At the same time, the CO2 concentration reaches up to 67%, and the CRR is about 70%. This rule can guide the design of an efficient polygeneration system cogenerating DRI and H2.

5. CONCLUSION

This paper proposes a novel polygeneration system with CO2 capture for DRI and H2 production. Through thermodynamic exergy analysis, it concludes that there is a large potential optimization in both physical and chemical energy utilization for the single DRI production system, and the proposed polygeneration system has a better thermodynamic performance than the single production systems. Besides, the polygeneration system achieves low energy penalty for CO2 capture.

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REFERENCE

[1] Li X, Fan TJ, Zhang LQ. China iron and steel industry energy saving and low carbon development report (2018) [R]. Beijing, 2018.

[2] Noriaki M, Takashi K, Norihide M. Application of coke oven gas to Midrex process[J]. Kobelco Technology Review. 2015;33:16-20.

[3] Liu BN, Li Q, Zou ZS, et al. Discussion on chemical energy utilisation of reducing gas in reduction shaft furnace[J]. Ironmaking & Steelmaking, 2014, 41(8): 568-574.

[4] Yi L, Huang Z, Peng H, et al. Action rules of H2 and CO in gas-based direct reduction of iron ore pellets[J]. Journal of Central South University, 2012, 19(8): 2291-2296.

[5] Benique FB, D'Abreu JC, Otaviano MM, et al. Kinetic of DRI carburization in direct reduction shaft furnaces[J]. Tecnologia em Metalurgia, Materiais e Mineracao, 2011, 20110912: 379-388.

[6] Boechat FO, De Carvalho RM, Tavares LM. Simulation of mechanical degradation of iron ore pellets in a direct reduction furnace[J]. Kona Powder and Particle Journal. 2018:216-25.

[7] Sarkar S, Bhattacharya R, Roy GG, et al. Modeling Midrex based process configurations for energy and emission analysis[J]. Steel Research International, 2018, 89(2): 1700248.

[8] Likkasith C, Saebea D, Arpornwichanop A, et al. Simulation of hydrogen production with in situ CO2 removal using AspenPlus[J]. Chemical Engineering Transactions, 2014, 39: 415-420.