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Invited Review

Development and progress on hydrogen metallurgy

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Abstract: Hydrogen metallurgy is a technology that applies hydrogen instead of carbon as a reduction agent to reduce CO_2 emission, and the use of hydrogen is beneficial to promoting the sustainable development of the steel industry. Hydrogen metallurgy has numerous applications, such as H_2 reduction ironmaking in Japan, ULCORED and hydrogen-based steelmaking in Europe; hydrogen flash ironmaking technology in the US; HYBRIT in the Nordics; Midrex H_2^{TM} by Midrex Technologies, Inc. (United States); H_2FUTURE by Voestalpine (Austria); and SALCOS by Salzgitter AG (Germany). Hydrogen-rich blast furnaces (BFs) with COG injection are common in China. Running BFs have been industrially tested by AnSteel, XuSteel, and BenSteel. In a currently under construction pilot plant of a coal gasification–gas-based shaft furnace with an annual output of 10000 t direct reduction iron (DRI), a reducing gas composed of 57vol% H_2 and 38vol% CO is prepared via the Ende method. The life cycle of the coal gasification–gas-based shaft furnace–electric furnace short process (30wt% DRI + 70wt% scrap) is assessed with 1 t of molten steel as a functional unit. This plant has a total energy consumption per ton of steel of 263.67 kg standard coal and a CO_2 emission per ton of steel of 829.89 kg, which are superior to those of a traditional BF converter process. Considering domestic materials and fuels, hydrogen production and storage, and hydrogen reduction characteristics, we believe that a hydrogen-rich shaft furnace will be suitable in China. Hydrogen production and storage with an economic and large-scale industrialization will promote the further development of a full hydrogen shaft furnace.

Keywords: hydrogen; hydrogen metallurgy; blast furnace; shaft furnace; low carbon

1. Introduction

According to the International Energy Agency (IEA), the total global energy-related CO_2 emission in 2018 was 3.31 billion tons, which increased by 1.7% compared with that in the previous year and reached the highest level on record. The CO_2 emissions of China, the US, Europe, and India were 9481, 4888, 3956, and 2999 Mt, respectively, accounting for more than 60% of the world's total CO_2 emissions [1]. Industrial production contributed 33% and 40% of the global energy consumption and CO_2 emissions, respectively. Furthermore, CO_2 emissions from the steel industry constituted a high proportion equivalent to about 33.8% of industrial emissions [2].

The Paris Agreement was adopted in 2015 to mitigate global warming. Its main goal is to limit the rise in global average temperature to less than 2°C in this century and to keep the global temperature rise to less than 1.5°C above the preindustrial level. Several countries signed the agreement and proposed plans to reduce carbon emission. In 2016, China launched 10 low-carbon demonstration zones, 100 mitigation projects, and 1000 training opportunities for climate change to reduce the CO_2 emission per unit of GDP by 60%–65% by 2030 [3–4]. In 2017, China also established a carbon emission trading system [2] mainly targeting the steel industry; as such, mandatory CO_2 emission reduction would force steel enterprises to develop low-carbon technologies.

On the basis of global warming and energy transformation, governments prioritize the development and utilization of carbon-free or low-carbon energy. Hydrogen energy, with its diverse sources, low-carbon emission, high efficiency, and wide application range, has been regarded as the most promising clean energy in the 21st century and listed in the national energy strategic deployment by many countries [5–6]. Xu *et al.* [7] put forward an idea of iron ore reduction by hydrogen in 1999. In 2002, Xu [8] further conceptualized hydrogen metallurgy. In 2018, Gan, who also pointed out that the 21st century is the era of hydrogen, defined hydrogen metallurgy as a process by which water is generated through hydrogen reduction instead of carbon reduction with an extremely fast reaction rate and without emission [9]. Hydrogen metallurgy mainly includes blast furnace (BF) ironmak-

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ing with the addition of hydrogen and the production of direct reduction iron (DRI) by hydrogen [10–15]. In the present work, the development and progress on hydrogen metallurgy abroad and in China and the challenges and opportunities in hydrogen metallurgy are systematically discussed.

2. Development and progress on hydrogen metallurgy abroad

2.1. Hydrogen metallurgy in Asia

2.1.1. Hydrogen reduction ironmaking in Japan

The route of the COURSE50 project shown in Fig. 1 involves two main technologies of hydrogen-rich reduction and CO_2 capture and recovery from a BF exhaust to achieve CO_2 emission. The former is based on the reforming technology of coke oven gas (COG) and water and a novel coking technology to make coke with a high strength and a high reactivity. The latter is based on an efficient CO_2 absorption technology to utilize waste heat. Nippon Steel built a 12 m^3 experimental BF with a productivity of 35 t/d and confirmed the project emission reduction target of 10% reduction by hydrogen reduction ironmaking and 20% reduction by CO_2 recovery; thus, the overall emission reduction target is 30% [16–17].

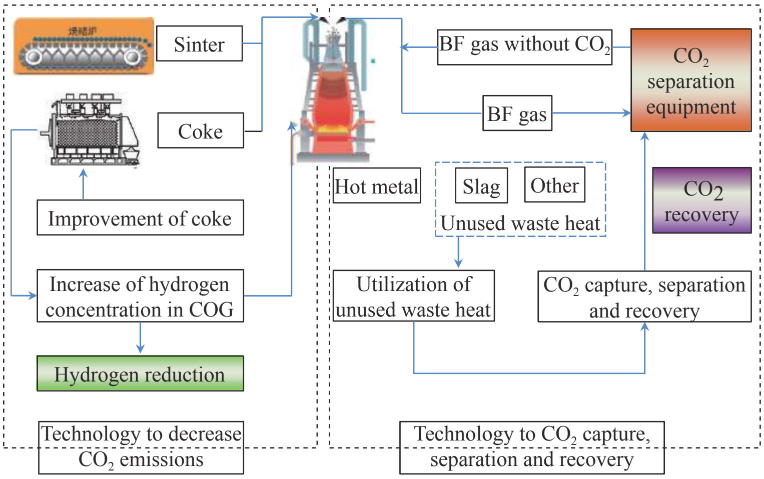

Fig. 1. Route of the COURSE50 project. Reprinted from *Energy Procedia*, 37, S. Tonomura, Outline of Course 50, 7160-7167, Copyright 2013, with permission from Elsevier.

In hydrogen reduction ironmaking, a portion of coke is replaced with hydrogen to reduce CO_2 emissions during BF process. The first stage of an experimental BF operation with H_2 injection carried out in 2014–2016 indicated that the carbon emission is reduced by 9.4% compared with that of non-H_2 injection. The adjustable position in the stack and raceway of BF and the preheating of H_2 before injection are conducted to ensure the maximum reduction performance and a stable inner temperature because of the small density of H_2 and endothermic reactions accompanying hydrogen reduction. At the second stage, an expanded test is carried out to gradually simulate a practical BF of 4000–5000 m^3. The first BF will be operated with H_2 reduction by 2030, and the technology will be available in Japan by 2050. The COURSE50 project has also been developed to produce hydrogen from COG. When COG leaves a carbonization chamber, the temperature reaches 800°C, which can maximize the use of sensible heat to catalytically crack tar and hydrocarbon substances and consequently produce H_2. The industrial trial of this technology has been completed. Through this modification, the H_2 content of COG increases from 55vol% to 63vol%–67vol%, and the gas volume doubles [18].

According to the 12th CSM Steel Congress report by Inoue [19], who is the vice president of Nippon Steel Co., COURSE50 is the first step, and a series of relevant hydrogen metallurgy technologies, such as H_2 reduction in BF (internal H_2 and external H_2), H_2 reduction without BF, CO_2 capture and storage (CCS), and CO_2 capture utilization (CCU), have been undertaken or planned in Japan.

2.1.2. Hydrogen metallurgy projects in Republic of Korea

Hydrogen reduction ironmaking developed through public–private cooperation in Republic of Korea involves three steps: start of the trial operation in a test furnace in 2025; production of two BFs in 2030 to decrease CO_2 emission by 1.6%; complete hydrogen reduction ironmaking in 12 BFs in 2040 to further decrease CO_2 emission by 8.7% [20].

2.2. Hydrogen metallurgy in Europe

2.2.1. ULCORED and hydrogen-based steelmaking in ULCOS project

Ultra-low CO_2 steelmaking (ULCOS) is a project jointly initiated by 15 European countries and 48 enterprises to achieve at least 50% reduction in CO_2 emission. The overall route is shown in Fig. 2 [21]. The triangular matrix shows that reducing agent and fuel consumption can be reduced by considering three factors, namely, carbon, hydrogen, and electricity. This ternary figure also presents all existing energy structures, where coal is close to carbon on a carbon–hydrogen line, natural gas (NG) is near hydrogen, and H_2 is produced through water electrolysis on the hydrogen-electricity line. Five processes are mainly promoted in ULCOS: top gas recycling of BF (TGR-BF), direct reduction (ULCORED), smelting reduction (HISARNA), electrolytic iron ore process (ULCOWIN/ULCOLYSIS), and hydrogen-based steelmaking [21].

In an ULCORED process, the traditional reducing agent of coke is replaced with a coal gasification gas or NG, and the consumption of NG can be cut down by top gas recycling and gas preheating. In addition, the application of NG with partial oxidation helps avoid the use of a coke oven and reforming equipment to greatly reduce investment requirements. The ULCORED routes are shown in Fig. 3. For example, the charge in a ULCORED with NG is loaded from the top of the shaft furnace, and the exhaust after purification and NG are mixed and injected into the shaft furnace to react with the iron-bearing burden. The tail gas of the new process only contains CO_2, which can be stored through CCS. The CO_2 emission via the ULCORED process combined with CCS technology can be reduced by 70% compared with the average emission of BFs in Europe [22–25].

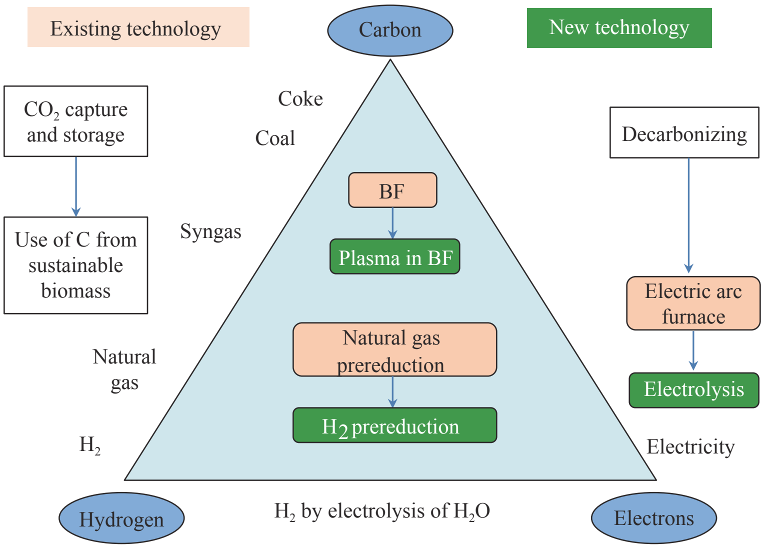

Fig. 2. Route of ULCOS. Reprinted from *Renewable Sustainable Energy Rev.*, 55, M. Abdul Quader, S. Ahmed, S.Z. Dawal, and Y. Nukman, Present needs, recent progress and future trends of energy-efficient Ultra-Low Carbon Dioxide (CO_2) Steelmaking (ULCOS) program, 537-549, Copyright 2016, with permission from Elsevier.

Hydrogen-based steelmaking is a hydrogen ironmaking technology with H_2 as a reducing agent. H_2 is made through water electrolysis, and the tail gas is water. The overall process is shown in Fig. 4 [26]. The carbon emission of the shaft furnace is negligible; however, when carbon emission generated by electric power is considered, CO_2 emission during the whole process per ton of crude steel is only 300 kg, and this value is equivalent to 84% reduction compared with that of 1850 kg from a traditional BF process [21–26].

2.2.2. H_2FUTURE by Voestalpine

The H_2FUTURE project initiated by Voestalpine in 2017 aimed to reduce CO_2 emissions from steel production by developing a breakthrough technology that could replace coke with hydrogen and thus ultimately reduce CO_2 emissions by 80% in 2050. This project also aimed to build the largest pilot hydrogen reduction plant around the world [27–28].

2.2.3. SALCOS project of Salzgitter AG

In 2019, Salzgitter low CO_2 steelmaking (SALCOS) was proposed by Salzgitter AG and Tenova to take hydrogen as a reducing agent in ironmaking and reduce CO_2 emissions. This project is designed to gradually transform a conventional and carbon-intensive BF conversion process to a DRI–electric arc furnace process and use surplus hydrogen for multiple purposes. In Green Industrial Hydrogen (GrInHy) 1.0, which was facilitated in 2016, a reversible electrolysis process is used to produce hydrogen and oxygen, and excess hydrogen is stored. Subsequently, GrInHy 2.0 was launched in 2019. The remarkable feature of GrInHy 2.0 is the use of waste heat from a steel plant to produce water vapor, which is applied to generate electricity from green renewable energy. Hydrogen is then produced via high-temperature water electrolysis. Hydrogen can be utilized in DRI production and later steel production processes, such as reducing gas for cold rolling annealing [29–31].

2.2.4. Replacement of coal with hydrogen by ThyssenKrupp Stahl AG

ThyssenKrupp Stahl AG initiated a project that involves the replacement of coal with hydrogen as a reducing agent in

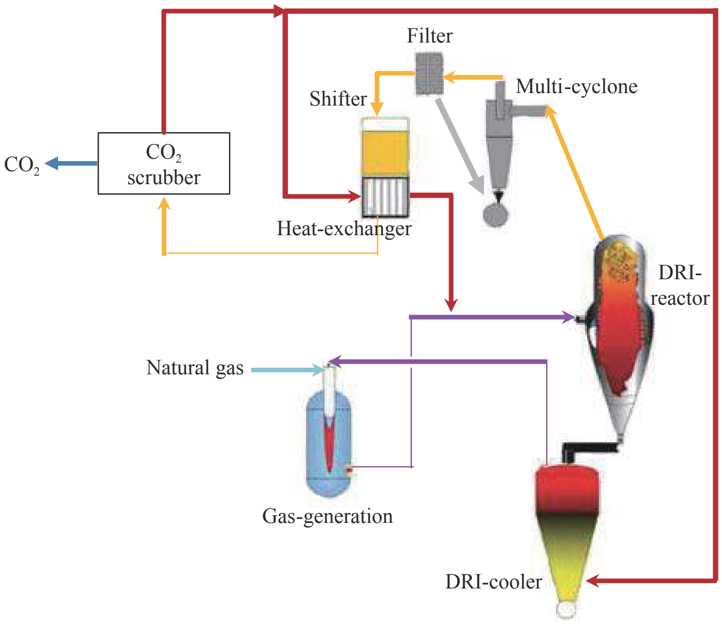

Fig. 3. Routes of ULCORED. From K. Meijer, M. Denys, J. Lasar, J.P. Birat, G. Still, and B. Overmaat, *Ironmaking Steelmaking*, 36(2009), No. 4, p. 251. With permission.

BF to reduce or completely avoid CO_2 emission in steel production. At the initial stage, hydrogen is injected to No. 9 BF through a tuyere in a Duisburg steel plant and gradually expanded to all 28 tuyeres. This technology will be installed in three other BFs in 2022. By then, the CO_2 emission from steel production will be decreased by about 20% [20].

2.2.5. COG injection system for ROGESA BFs

ROGESA Roheisengesellschaft Saar mbH (ROGESA) has awarded Paul Wurth to design and supply COG injection systems for BF Nos. 4 and 5 located in Dillingen/Saar, Germany. COG partially replaces pulverized coal and metallurgical coke as reducing agents in the BF process. Thus, the carbon intensity in the BF and the carbon footprint during the overall ironmaking process are further decreased. This application will be a bold step toward hydrogen-based ironmaking in the future. According to the project schedule, COG is initially injected into half of the total number of the tuyeres of BF No. 5 in summer in 2020; by the end of 2020, COG is permanently injected into all the tuyeres of both furnaces [32].

2.3. Hydrogen flash ironmaking technology in the US

A novel flash ironmaking technology (FIT) involves the reduction of iron concentrate to DRI with a high metallization rate under a suspension state. The reducing gas can be H_2, CH_4, or other gases. This process is developed on the basis of iron ore resources in the US, where 60% of iron ore is taconite with a small particle size of less than 25–38 μm. In this process, iron oxide concentrates are directly used without pelletizing or sintering [33].

At the University of Utah, large-scale tests have been carried out on the basis of FIT by using an iron concentrate from ArcelorMittal and Ternium and a shaft furnace with an inner diameter of 20.3 cm and a length of 244 cm. A reduction degree of 90%–99% can be rapidly obtained within 1–7 s at 1200–1400°C, and this parameter depends on the excess coefficient of hydrogen. The comparisons of energy consumption between a BF process and FIT that involves H_2, CH_4, and coal via mathematical calculations are investigated. The energy consumption of FIT is remarkably lower than that of the BF process. For 1 t of iron, CO_2 emissions are 71, 650, and 1145 kg when H_2, CH_4, and coal are utilized in FIT, respectively. By comparison, the emission of conventional BF is as high as 1671 kg. Even if coal serves as the fuel, the CO_2 emission of FIT is significantly lower than that of the BF process [34–35].

2.4. Hydrogen breakthrough ironmaking technology (HYBRIT) in the Nordics

In 2016, Vattenfall, SSAB (a Swedish steel company), and LKAB (a Swedish mining company) co-founded a nonfossil energy project called HYBRIT. The core concept of this project is direct reduction by hydrogen made via water electrolysis, and its goal is to reduce CO_2 emissions by 10% in Sweden and 7% in Finland [36–39].

This HYBRIT new process is similar to direct reduction in existing shaft furnaces, but 100% hydrogen is used as a reducing agent. Base on the Swedish raw material and production data, the comparison of CO_2 emission and energy consumption between HYBRIT and the BF process for 1 t had been discussed. In the BF process, pelletizing, coking, ironmaking, and steelmaking are the main processes considered.

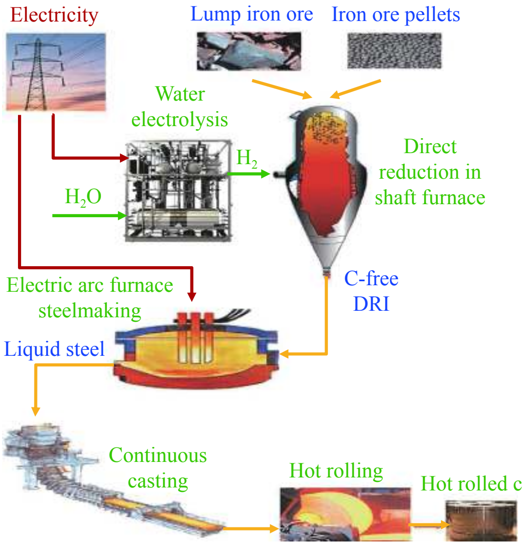

Fig. 4. Hydrogen-based steelmaking in ULCOS project. Reprinted from *J. Cleaner Prod.*, 46, A. Ranzani da Costa, D. Wagner, and F. Patisson, Modelling a new, low CO_2 emissions, hydrogen steelmaking process, 27-35, Copyright 2013, with permission from Elsevier.

CO_2 emission, fossil energy consumption, and electric power consumption are 1600 kg, 5231 kW·h, and 235 kW·h, respectively, and the total energy consumption is 5466 kW·h. In HYBRIT, the main processes are pelletizing, hydrogen production, direct reduction by hydrogen, and electric furnace smelting. CO_2 emission, biomass energy consumption, fossil energy consumption, and electricity consumption are 25 kg, 560 kW·h, 42 kW·h, and 3488 kW·h, respectively, and the total energy consumption is 4090 kW·h. The CO_2 emission in HYBRIT decreases by 1575 kg (98.44%), and its energy consumption reduces by 1356 kW·h (25.17%) compared with that in the BF process. Furthermore, a 10% reduction in CO_2 emission in Sweden leads to a 15 kW·h increase in power consumption, which can be achieved by wind power and solar power generation. Three pilot plants will be built: a hydrogen production–direct hydrogen reduction steelmaking plant in Lulea, a nonfossil pellet plant in Malmberg and Lulea, and a hydrogen storage plant in Lulea [36–39].

2.5. Direct reduction technology by hydrogen-rich gas

Since the mid-20th century, direct reduction by hydrogen-rich gas has been industrialized. At present, running shaft furnaces are mostly based on Midrex and Hylsa (HYL) processes by using NG, coal, and oil-gasified gas. The reducing gas should have a high hydrogen content of 55vol%–80vol% to ensure that a high-temperature alloy furnace tube is not corroded and to prevent iron ore pellet from sticking [40]. A Midrex hot briquetted iron (HBI) plant, which belongs to Voestalpine with an annual capacity of 2 million tons, was established in 2017. Nucor invested $750 million to build the first native HYL DRI plant with an annual capacity of 2.5 million tons in Louisiana in 2013 [41].

In Midrex, the contents of H_2 and CO in the reducing gas are about 55vol% and 36vol%, respectively, and the H_2/CO molar ratio is about 1.50. The H_2/CO molar ratio in the FMO Midrex plant in Venezuela is 3.3–3.8 because of steam reforming technology. Coal gasification technology is used in some shaft furnaces to obtain a reducing gas with a H_2/CO molar ratio of 0.37–0.38. Therefore, Midrex has been successfully applied to produce DRI at a H_2/CO molar ratio of 0.37–0.38. Midrex with pure hydrogen (i.e., Midrex H_2^{TM}) may be developed with a route based on Midrex with a H_2/CO molar ratio of 0.37–0.38. In Midrex-H_2^{TM}, reforming is eliminated, and hydrogen is heated directly to the required temperature. In practical production, the reducing gas contains about 90vol% hydrogen and 10vol% other gases (i.e., CO, CO_2, H_2O, and CH_4), which are mainly used to control the appropriate furnace temperature and ensure the carburization of DRI [42]. In 2019, ArcelorMittal announced that it commissioned Midrex Technologies to design a demonstration plant at its Hamburg site, produce steel with hydrogen, demonstrate large-scale production, and use DRI made of 100vol% hydrogen as a reductant. In the future, the demonstration plant will produce about 100000 t of DRI per year initially with gray hydrogen sourced from NG. Once green hydrogen from renewable energy sources becomes available in sufficient quantities and at an economical cost, hydrogen can be produced from wind farms off the coast of Northern Germany. The plant will be the world's first industrial-scale DRI production plant powered by hydrogen [43].

3. Development and progress on hydrogen metallurgy in China

Hydrogen reduction technology has been widely explored because of the increasing pressure to reduce CO_2 emission. Hydrogen use during BF and direct reduction processes has been discussed and facilitated in China. The fundamental conditions for hydrogen production from nuclear energy and hydrogen metallurgy have also been made available in China.

3.1. Development of hydrogen-rich BF in China

BF ironmaking is the core of energy saving and CO_2 reduction in the steel industry. Hydrogen-rich reduction is one of the feasible low-carbon ironmaking technologies. In hydrogen-rich BF processes in China, hydrogen-rich gases, such as COG and NG, are injected into a BF.

As early as the 1960s, a BF operation with COG injection was tested in Ben Steel. At a COG injection rate of 82 m^3 per ton hot metal the productivity increased by 10.8%, the coke ratio decreased by 3%–10%, the temperature of BF was stable, the phenomena of collapse burden and suspended burden enhanced, and the condition of BF improved. Combining the experience of injecting COG into the BF process in Ben Steel, An Steel conducted the No. 9 BF operation with COG injection in 1964; for 1 m^3 of COG injection, 0.6–0.7 kg of coke could be saved. In 2012, An Steel started the COG injection in No. 1 BF in Bayuquan. The annual economic benefit was about 100 million yuan, and the annual reduction of CO_2 emission was approximately 650000 t [44].

3.2. Development of hydrogen metallurgy direct reduction in China

The global development trend of DRI has shown that gas-based shaft furnace processes are effective in rapidly expanding DRI. Since the end of the 20th century, studies have been conducted on gas-based shaft furnace direct reduction approaches, such as an industrial test on the Baosteel and Lunan Chemical Industry Group (BL) coal gasification–shaft furnace process at Baosteel, a semi-industrial test on coal gasification–shaft furnace process at a Hengdi plant, and a COG–shaft furnace at Zhongjin Mining.

With the development of coal gasification in the chemical industry and the progress on shaft furnace direct reduction, coal gasification–shaft furnace processes have been extensively investigated. Large-scale development of gas-based direct reduction technology is unsuitable in China because of the shortage and high price of oil and NG and the domestic energy structure. However, China is rich in coal resources, especially noncoking coal resources. The combination of coal gasification and direct reduction is the main development direction for the steel industry in China. Moreover, the reducing gas generated from coal gasification technology is rich in H_2 (about 60vol%). Therefore, coal gasification–shaft furnace direct reduction is an important part of hydrogen metallurgy.

3.2.1. Key integration technologies of coal gasification–shaft furnace direct reduction

The key integration technologies of coal gasification–shaft furnace process mainly include iron concentrate dressing, special oxidized pellet preparation, optimization for a gas-based shaft furnace, reasonable coal gasification selection, gas-based reduction control, and energy utilization improvement.

Tang *et al.* [45] reported that a high-grade iron concentration can be economically produced with total Fe and SiO_2 contents of more than 70.5wt% and less than 2.0wt% through grinding and magnetic separation based on magnetite resources in China; these conditions can be directly applied to prepare excellent oxidized pellets.

Wang [46] revealed that oxidized pellets with excellent metallurgical properties can be obtained by optimizing the use of additives and improving the technological parameters of drying, preheating, and oxidation roasting. Comparisons between the performance levels of pellets prepared from a domestic iron concentrate and HYL indices are listed in Table 1. The performance levels can satisfy the requirements of the HYL process. The shaft furnace with a high temperature and a high hydrogen content is adopted. The qualified DRI with a metalized rate of not less than 92% and a SiO_2 content of not more than 3wt% can be obtained by controlling the reduction process. A comprehensive weighted scoring method based on the comprehensive considerations of equipment characteristics, the technical and economic indices, and investment is applied to evaluate the rational selection of coal gasification technologies. A fluidized bed method with advantages of low investment, low oxygen consumption, and high production efficiency is a proper choice. Furthermore, a hot delivery with top gas recycling likely enhances the energy utilization of a gas-based shaft furnace process.

3.2.2. Development of a pilot plant of coal gasification–gas-based shaft furnace co-built by Northeastern University and Liaoning Huaxin

In 2018, Liaoning Iron and Steel Generic Technology Innovation Center was established by Northeastern University and Liaoning Huaxin to build a national demonstration of low-carbon ironmaking and high-quality steelmaking based on a short hydrogen metallurgical process. The important content of this project is a coal gasification–shaft furnace with an annual DRI of 10000 t. The main route is shown in Fig. 5.

Table 1. Comparisons between the performance levels of pellets prepared from domestic iron concentrate and HYL indices

	Chemical composition / wt%			Physical properties		Metallurgical properties				
Concentrate	TFe	FeO	SiO_2	CS / N (GB/T 14201)	DS / % (GB/T 8209)	RDI / % (GB/T 13242)			RD / % (GB/T 13241)	RSI / % (GB/T 13240)
						$RDI_{+6.3}$	$RDI_{+3.15}$	$RDI_{-0.5}$		
Jilin	67.30	0.10	2.65	3276	94.5	87.5	91.4	6.5	72.83	18.70
Shanxi	67.90	0.19	1.72	2985	94.4	95.7	97.0	2.4	85.71	16.78
Liaoning	68.36	0.19	2.11	2598	94.7	80.0	84.2	6.5	69.90	17.73
HYL index	As high as possible	≤1.0	≤3.0	≥2000	≥93	—	—	—	—	≤20

Note: CS—Compressive strength; DS—Drum strength; RDI—Reduction degradation index; RD—Reduction degree; RSI—Reduction swelling index.

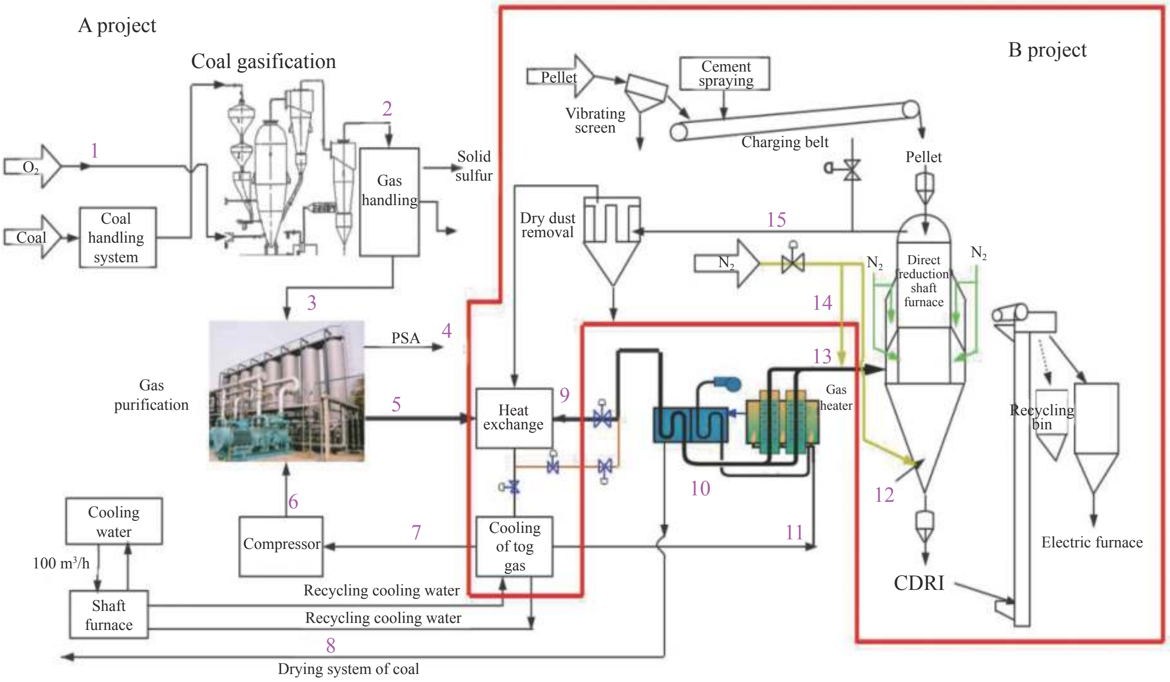

Fig. 5. Coal gasification–gas-based shaft furnace of Northeastern University and Liaoning Huaxin. 1—O_2; 2—Crude gas of coal gasification; 3—Crude gas after desulfuration; 4—Tail gas after SPA; 5—Purified gas; 6—Pressed top gas; 7—Top gas after cleaning and drying; 8—Exhaust after combustion; 9—Heat exchange gas; 10—Combustion air; 11—Heating gas for heat furnace; 12—Cooling NG; 13—Input gas; 14—N_2; 15—Hot top gas.

The Ende method has been adopted to gassify coal. In this method, raw coal must be noncohesive or weakly cohesive, the content of ash must be less than 25wt%–30wt%, and the chemical activity must be good. The contents of H_2, CO, and CH_4 in the clean gas obtained by the Ende method with Huolinghe coal as a raw material are 57vol%, 38vol%, and 2vol%, respectively. One Ende furnace with a gas production capacity of 5000 m^3/h should be adopted on the basis of material and energy balances.

According to production requirements, the following parameters are calculated: effective gas flow, reduction temperature, utilization coefficient of shaft furnace, burden charge, and other key parameters, such as proper shaft diameter, stack angle, and heights of reduction, transitional, and cooling parts. The shaft furnace with an independent intellectual property right has also been developed. The three-dimensional model and final assembly diagram of this gas-based shaft furnace are shown in Fig. 6. First, the multifield and multiphase coupling simulations of gas flow reduction, reduction process, burden movement, and temperature field are carried out on the basis of domestic raw materials. The distribution of a solid phase during gas-based reduction ($Fe_2O_3 \rightarrow Fe_3O_4 \rightarrow FeO \rightarrow Fe$) is shown in Fig. 7. Second, the process models of each unit, including coal gasification, pelletizing, gas-based reduction, and electric furnace smelting, are established. The material balance and exergy balance of a shaft furnace are presented in Tables 2 and 3, respectively. The highest exergy input is the chemical exergy of reducing gas, and the exergy of the top gas accounts for 57% of the output. Therefore, the improvement of the utilization of the top gas is the key to enhance the energy utilization of gas-based shaft furnace processes. Lastly, GaBi 7.3 software is used to conduct an overall life cycle assessment (LCA) of the coal gasification–shaft furnace–electric furnace process (30wt% DRI + 70wt% scrap) with 1 t of molten steel as the functional unit. The overall environmental impact of

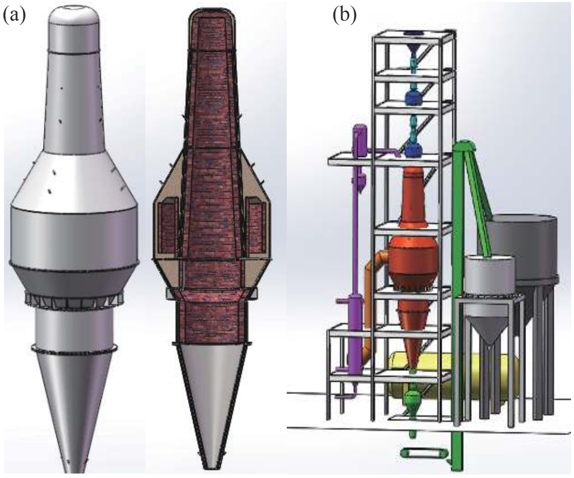

Fig. 6. (a) Three-dimensional models and (b) final assembly diagram of a gas-based shaft furnace.

this system is 1.83E-11, and the three environmental impact types that make the highest contribution are as follows: climate change (GWP100, 48.39%), photochemical ozone creation potential (POCP, 39.86%), and eutrophication (EP, 7.24%). The key processes that have the greatest impact on the environment are electric furnace smelting, gas heating, and coal gasification. Under these conditions, the total energy consumption per ton of steel is 263.67 kg standard coal, and the CO_2 emission per ton of steel is 829.89 kg. These results are superior to those of a traditional BF process.

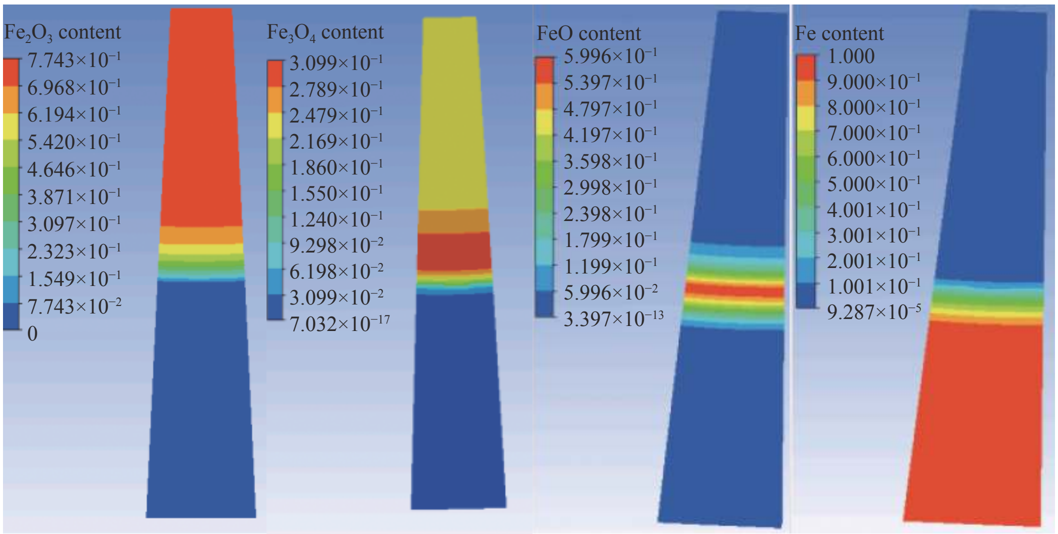

Fig. 7. Solid phase concentration distribution during reduction ($Fe_2O_3 \rightarrow Fe_3O_4 \rightarrow FeO \rightarrow Fe$).

Table 2. Material balance of the shaft furnace used kg

Input			Output			
Pellet	Reducing gas	Total input	DRI	Top gas	Ash	Total output
1377.85	647.88	2025.73	1000.00	1011.95	13.78	2025.73

Table 3. Exergy (Ex) balance of the shaft furnace used

Item	Exergy type	Exergy / MJ	Ratio / %
Input	Pellet chemical Ex	21.39	0.11
	Reducing gas physical Ex	1182.92	6.34
	Reducing gas chemical Ex	17446.37	93.54
	Total input Ex	18650.69	100.00
Output	DRI physical Ex	186.29	1.00
	DRI chemical Ex	6161.96	33.04
	Ash physical Ex	1.69	0.01
	Ash chemical Ex	0.21	0.00
	Top gas physical Ex	469.36	2.52
	Top gas chemical Ex	10730.26	57.53
	Heat loss Ex	130.23	0.70
	Inner loss Ex	970.69	5.20
	Total output Ex	18650.69	100.00

4. Challenges and opportunities in hydrogen metallurgy

4.1. Hydrogen production and storage

The key factors of hydrogen production are economy and low carbon. Fossil fuel energy is mainly applied to produce hydrogen, which accounts for more than 95% of hydrogen in the world. However, a large amount of CO_2 is generated by this technology. In the long run, technologies based on solar energy, wind energy, water energy, marine energy, and geothermal energy will emerge as important supplement resources for hydrogen production to realize zero-carbon emission. The conversion efficiency of these technologies is still relatively low, but hydrogen production from solar energy has been used as a temporary and supplementary hydrogen refueling process in Europe and Japan. With the improvement of CCS technology, the combination of hydrogen production from coal gasification and CCS technology will provide a new way to cleanly and efficiently utilize coal resources; this combination is also the development direction of coal-based and low-carbon hydrogen production [47–48].

Hydrogen storage technology is a restrictive step to efficiently utilize hydrogen. Currently, hydrogen storage methods mainly included high-pressure gaseous storage, cryogenic liquefied storage, organic and liquid storage, porous material storage. For the large-scale storage and transportation of hydrogen energy, the most feasible technologies in the industry are high-pressure gaseous storage and cryogenic liquefied storage, although many technologies have been developed [47–48].

4.2. Problems on gas-based shaft furnaces with pure hydrogen

Since the industrial production in the 1980s in Western Europe, shaft furnaces with pure hydrogen have been considered an important development direction of hydrogen metallurgy. Although many authoritative metallurgical experts emphasized the benefits of hydrogen metallurgy, the technological and economic value of shaft furnaces with pure hydrogen should still be considered and discussed.

No carbon source is found in a pure hydrogen shaft furnace, leading to the impossible complementation and transformation of heat and the recycling of substances. In a shaft furnace with pure hydrogen, three strong endothermic reactions occur.

$$Fe_2O_3 + 3H_2 \rightarrow 2Fe + 3H_2O \quad (1)$$

$$Fe_3O_4 + 4H_2 \rightarrow 3Fe + 4H_2O \quad (2)$$

$$FeO + H_2 \rightarrow Fe + H_2O \quad (3)$$

Therefore, a large amount of heat is absorbed, and the inner temperature at the bulk material layer decreases rapidly. As a result, the reduction reactions that need to consume a high amount of heat and worsen the gas utilization rate are delayed. The amount of hydrogen as a heat carrier should be increased to maintain the preferred productivity. For example, when the pressure at the top is 0.4 MPa, the amount of hydrogen with a temperature of 900°C should be at least 2600 m^3 per ton direct reduction iron (DRI) to meet the heat demand of shaft furnace reduction. If the hydrogen supplement remains unchanged, the DRI output is one-third less than that of the current, leading to a great increase in DRI cost.

The specific gravity of hydrogen is low, and the density of hydrogen is only 1/20 of that of CO. As a result, the entering hydrogen molecule rapidly escapes upward. In comparison with the path and direction of a mixed reducing gas, those of hydrogen in a furnace change so quickly that hydrogen cannot stay in the high-temperature zone at the bottom part of the shaft furnace to complete the task of reducing iron ore pellets. Theoretically, DRI products can also reach the designed index by maintaining the entering hydrogen with a pressure above 1 MPa and a temperature above 1000°C. However, hydrogen is an extremely flammable and explosive substance, and the shaft furnace needs highly efficient and long-term stable production. If the shaft furnace system is allowed to work for a long time under the ultimate conditions of high temperature and high pressure, safety cannot be guaranteed, and this issue is not in line with the goal of metallurgical process designs.

Hydrogen is an expensive secondary energy. Profitable and commercialized DRI production in a shaft furnace with pure hydrogen is difficult to be achieved. Moreover, the cost of a shaft furnace with pure hydrogen, including equipment, investment and maintenance, is nearly twice as high as that of current coal gasification–gas-based shaft furnaces. Furthermore, a small amount of CO is found in the running shaft furnaces of Midrex, HYL, and PERED. Exothermic and endothermic reactions are carried out simultaneously in different parts of furnaces; consequently, the heat exchange and the thermodynamic and dynamic conditions of heat and mass transfer in shaft furnaces improve.

In summary, direct reduction rate and production efficiency are affected by many factors, such as hydrogen proportion, temperature, pressure, gas utilization rate, residence time of iron ore, heat transfer, mass transfer, and shaft furnace design. The developed coal gasification technology and the production and storage technology of hydrogen should be improved on the basis of abundant coal resources.

With the progresses of relevant technologies, the law of countercurrent reduction shaft furnace is obeyed; reliable, stable, efficient, and energy saving methods are introduced; the advantages of rapid reaction rate and mass transfer speed are promoted; the existing mature industrialization of shaft furnace designs and production operation experiences are fully used; key equipment is improved; the process parameters of shaft furnace reduction are optimized; the advantages of hydrogen-rich shaft furnaces are maximized; and proper capacity and low energy consumption are obtained. Then the use of a hydrogen-rich shaft furnace would be more easily successful than that of a shaft furnace with pure hydrogen in China. Meanwhile, with the development and practice of zero-emission, economic and large-scale hydrogen production based on solar energy, wind energy, water energy, marine energy, and geothermal energy, as well as the development and industrialization of hydrogen storage devices and technologies, pure hydrogen metallurgy can be achieved.

5. Conclusions

(1) In hydrogen metallurgy, hydrogen is applied as a reducing agent instead of carbon to cut down CO_2 emission and promote the green and sustainable development of the iron and steel industry. Several hydrogen metallurgical projects or methods abroad are mainly given as follows: H_2 reduction ironmaking in Japan, ULCORED and hydrogen-based steelmaking in Europe, hydrogen FIT in the US; HYBRIT in the Nordics; Midrex H_2^{TM} by Midrex Technologies, Inc. (United States); H_2FUTURE by Voestalpine (Austria), SALCOS by Salzgitter (Germany); and replacement of coal with hydrogen by ThyssenKrupp Stahl AG (Germany).

(2) COG injection is typical in hydrogen-rich BF smelting processes in China. Some steel plants, such as Ben Steel, An Steel, and Mei Steel, have conducted BF operations with COG injection and gained some positive effects. At a COG injection rate of 82 m^3 per ton hot metal in Ben Steel, productivity increases by 10.8%, the coke ratio decreases by

3%–10%, and the BF condition improves.

(3) The following key technologies of a coal gasification–shaft furnace process are available in China: iron concentrate dressing, special oxidized pellet preparation for a gas-based shaft furnace, coal gasification technology, gas-based controlling reduction technology, and energy utilization improvement. In a currently under construction pilot plant of a coal gasification–gas-based shaft furnace with an annual DRI output of 10000 t, the reducing gas composed of 57vol% H_2 and 38vol% CO is prepared via the Ende method. With 1 t of electric furnace steel as a functional unit, the LCA of the coal gasification–gas-based shaft furnace–electric furnace short process is conducted. The results reveal that three types of environmental impacts mostly contribute to this process: GWP100 (48.39%), POCP (39.86%), and EP (7.24%). Under these conditions for each ton of steel, the total energy consumption is 263.67 kg standard coal, and the CO_2 emission is 829.89 kg, which are both superior to a traditional BF process.

(4) The equipment and technology of hydrogen production and storage need further improvement. Meanwhile, the endothermic hydrogen reduction reduces the temperature of the shaft furnace, the specific gravity of hydrogen is small, and the cost of hydrogen production is high. Therefore, a hydrogen-rich shaft furnace should be developed using raw materials and fuels in China. The large-scale, zero-emission, and economic industrialization of hydrogen production and storage will promote the further development of a full hydrogen shaft furnace.

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