

High-Temperature Electrolysis Integrated with Direct Reduced Iron Process for Producing Low-Carbon Steel

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ABSTRACT

High-temperature electrolysis (HTE) of steam is one of the most energy-efficient techniques for producing hydrogen. Coupling the process with a direct reduced iron (DRI) plant can help the steel industry lower its carbon footprint by using the low-carbon hydrogen as a reducing agent and fuel, instead of synthesis gas derived from either natural gas or coal. In this study we aim to predict the performance of an integrated DRI plant via process modelling and simulation. The integration of HTE significantly reduced the CO₂ intensity of steel production.

Keywords: Hydrogen, Electrolysis, Direct Reduced Iron, Process Modelling, Decarbonization

INTRODUCTION

Iron and steel industries generate about 7% of the CO₂ energy related emissions as referenced by the International Energy Agency.¹ Most of the companies of the sector have thus made announcements to reach net zero between 2045 to 2050. Accordingly, the iron and steel industry decarbonization path² will at least comprise: (i) steel processes and materials optimization, (ii) developing CO₂ capture technologies and CO₂ utilization options, and (iii) feasible and economic low carbon hydrogen production. In addition, this technology and innovation journey will be combined with proper digital tools for measurements and evaluation purposes.

Manufacture of steel involves several high-temperature processes: (i) conversion of iron ore into steel, (ii) ladle refining and casting into multiple forms, (iii) heat treatment (e.g., quenching, annealing, and tempering) and (iv) chemical processes (e.g., galvanization). Among them, the direct reduced iron (DRI) process is one of the major steps of the steel making chain. The DRI allows to convert iron ore into sponge iron using a reducing agent such as synthesis gas (mixture of hydrogen and carbon monoxide) derived from either natural gas or coal. Pure hydrogen is also a fair option as reducing agent.³

Hydrogen is usually produced by methane reforming and coal gasification. Low-carbon hydrogen can also be produced by electrolysis technologies using water and renewable energy sources (such as solar, wind, hydraulic, and biomass). High-temperature electrolysis (HTE) of steam is one of the most energy-efficient techniques for producing hydrogen.⁴

In this work, we present both modelling and simulation of a DRI process based on the Energiron ZR scheme.⁵ A detailed process model of DRI is useful to predict the future performances of the plant combined with a low-carbon hydrogen supply from an HTE. Switching the conventional reducing agent supply by an electrolytic low-carbon hydrogen, impacts indeed the process operating conditions. The decarbonized scheme considers suitable connections of the conventional DRI process with the high temperature electrolysis system by mean of two interfacing units i.e., a heat recovery process (recovering waste heat from the DRI process to produce useful steam) and a hydrogen delivery system (supplying hydrogen at the appropriate specifications towards the DRI process).

The models were created using the SLB proprietary computational thermodynamic process modelling software platform, Symmetry®. Process simulations enabled determining the overall heat and mass balances and evaluating energy efficiency and direct carbon emissions. The main process parameters were also compared for the conventional and decarbonized methods.

PROCESS MODELLING, CALIBRATION AND VALIDATION

The compounds considered in the models are water, hydrogen, oxygen, nitrogen, methane, carbon dioxide, carbon monoxide, iron, iron monoxide, diiron trioxide, and triiron tetraoxide. Their thermodynamic properties are given by the Advanced Peng Robinson equation of state and the Symmetry® Gasification 2010 thermodynamic model. All inlet streams to the model battery limits are ambient conditions (25°C and 1.01 bar). The models are based on pure thermodynamic approach.

Each model – i.e., direct reduced iron process, heat recovery unit, high temperature electrolyzer, and hydrogen processing unit – is described according to available real and/or literature input design and process data. Predefined blocks and objects are used for each process conditions changes. Specific parameterizations are performed according to the type of blocks or objects. Heat exchanger blocks are modelled as counter-current flow exchangers and are specified by temperature approach. Pump and compressor blocks are specified by their discharge pressure and efficiency. Separators and splitters are described by split factors. Reactor blocks are specified as either equilibrium or conversion type reactor.

1. Direct Reduced Iron Process

The DRI process is modelled as shown in **Figure 1**. Methane feeds the process and is mixed with rich hydrogen recycle stream and water via mixer blocks (**M2** and **M3**). The obtained stream is heated at the DRI specific temperature in a heat exchanger block (**HX1**). The heat is provided by means of combustion of a second methane stream with air in a burner block (**BRN1**) where the oxygen outlet stream fraction is specified. A controller block (**CN1**) calculates the suitable fuel flowrate to ensure the DRI specific temperature. The DRI is modelled with two successive reaction blocks (**DRI-REF** and **DRI**) where methane reforming and iron oxides reduction occurred. An additional oxygen stream is mixed with the inlet of the reaction block (**DRI-REF**) to enhance the reforming reaction. To simplify the iron oxides reduction reaction scheme, iron ore is defined as pure diiron trioxide. This latter feeds the reaction block (**DRI**) after heat exchange with the gaseous outlet stream of the DRI reaction in a feed-effluent heat exchanger (**HX2**). The gaseous outlet stream is then cooled down via cooler block (**C1**) to allow the water removal in (**SEP1**). The obtained lean water stream is split at splitter block (**SP2**). Part of this stream can be used as fuel in the burner block (**BRN1**), and the remaining is processed in a CO₂ separator (**CO2-CAPT**) to generate a rich hydrogen recycle stream. Iron is obtained at the outlet of the reaction block (**DRI**).

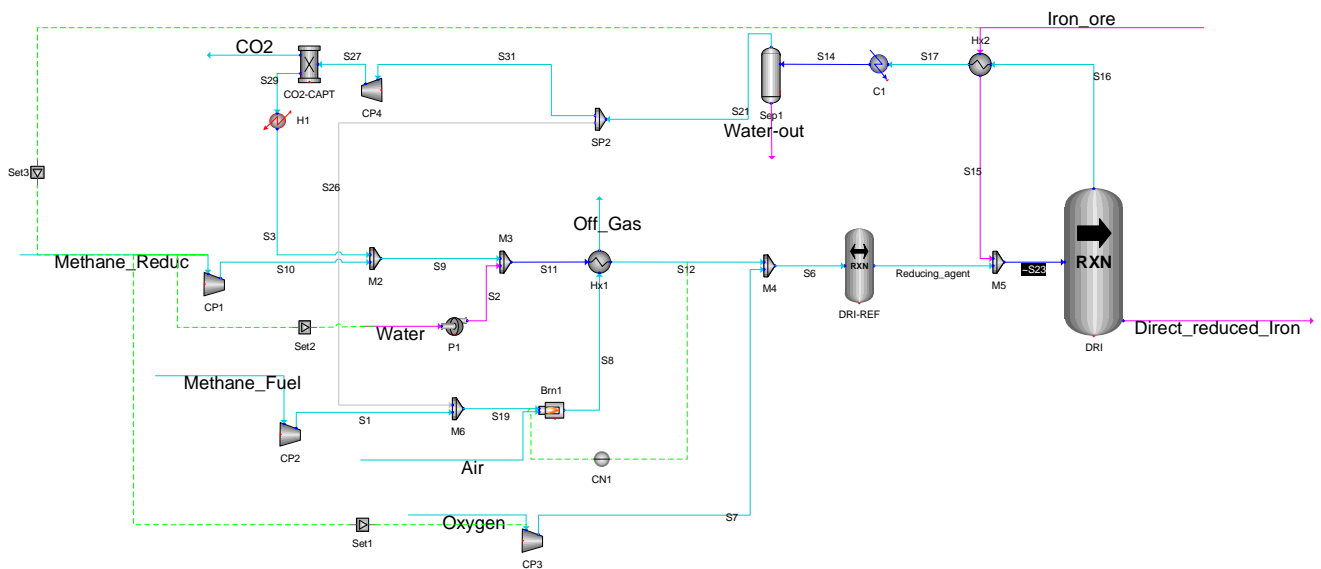


Figure 1. Symmetry® model of the conventional DRI process.

Switching from methane to hydrogen, implies some changes in the DRI model. The methane reforming reaction block (**DRI-REF**), the additional oxygen stream, and the CO₂ separator (**CO2-CAPT**) are removed.

2. Heat Recovery Unit

The heat recovery unit (HRU) is modelled as an indirect heat recovery steam generation system as shown in **Figure 2**. Hot source stream passes through an economizer type heat exchanger block (**ECON**) to exchange heat with a circulating thermal transfer fluid. The hot source is drove by mean of a fan modelled by a compressor block (**CP2**) and the circulation of the thermal transfer fluid stream is ensured by a pump block (**P3**). The obtained hot thermal transfer fluid feeds another heat exchanger block (**SG-SH**) representing the successive water preheating, water vaporization, and steam superheating. Water supply is realized via a feed water pump block (**FWP1**). Superheated steam leaves the HRU at the outlet of the heat exchanger block (**SG-SH**).

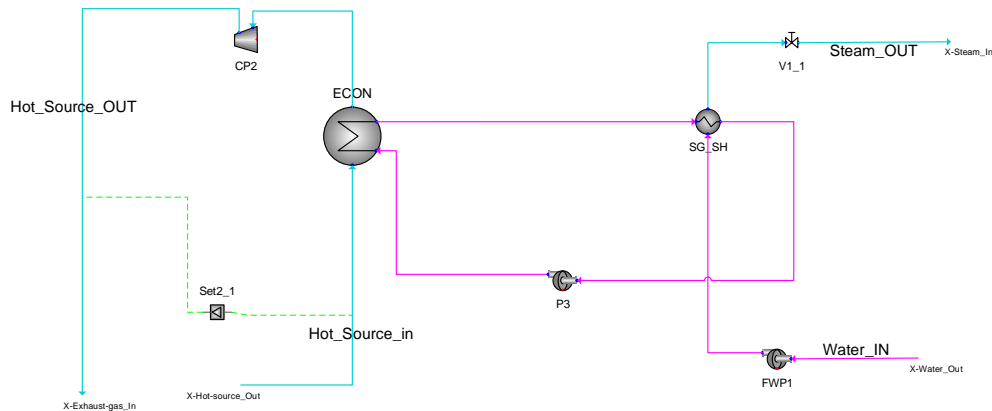


Figure 2. Symmetry® model of the HRU.

3. High Temperature Electrolyser

The high temperature electrolyzer (HTE) is modelled as shown in **Figure 3**. Steam feeds the process and is mixed with a recycle stream via a mixer block (**M1**). The obtained stream is heated at the electrolysis temperature in successive feed-effluent heat exchanger block (**E1**) and electric heater exchanger block (**H1**). Sweep gas also feeds the process and is heated in successive feed-effluent heat exchanger block (**E2**) and electric heater exchanger block (**H2**). Both hot steam and hot sweep gas enter the electrolysis cell stack (**ELEC1**) where the electrolysis reaction happened. The electrolysis cell stack (**ELEC1**) is specified by stack arrangements, electrode geometry, current density, cell potential, minimum overpotential, and system thermal and hydraulic performances. Oxygen molecules are formed at the anode side and is transported by the sweep gas. Hydrogen molecules are formed at the cathode and leave the electrolysis cell together with unreacted water. The mixture of hydrogen and water is then processed in an aero-refrigerant type heat exchanger block (**AC1**) and a cooler block (**C2**) to eliminate most of the unreacted water. The raw hydrogen is drove via a blower (**CP2**) and part of the stream is recycled at the HTE inlet to prevent oxidation of the stack. The remaining raw hydrogen exit the HTE unit.

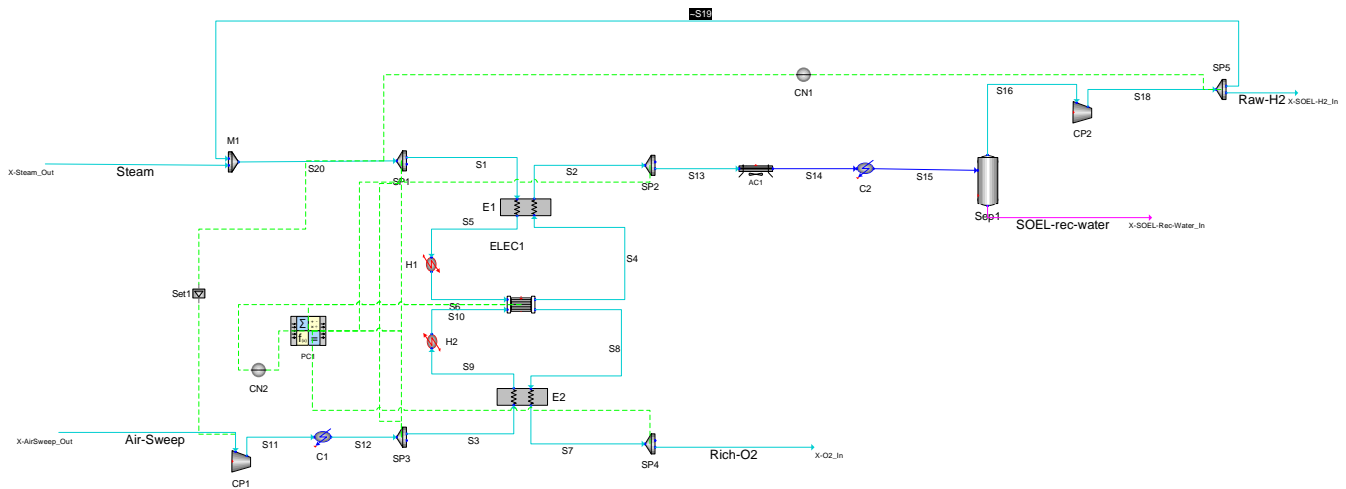


Figure 3. Symmetry® model of the HTE.

4. Hydrogen Processing Unit

The hydrogen processing unit (HPU) is modelled as shown in **Figure 4**. Raw hydrogen feeds the process and is first cooled down in a cooler block (**C1**) to eliminate water in a separator (**SEP1**). The obtained stream is optionally mixed with a recycle stream and compressed in a compressor block (**CP1**) to reach a specific pressure. The stream is then cooled down again in another cooler block (**C2**) to eliminate remaining water trace in another separator (**SEP2**). The rich hydrogen stream is at that stage at the required temperature and pressure for the purification via pressure swing adsorption. This latter is modelled by four adsorption /desorption block (**ADS1/2** and **DES1/2**) by specifying split factors. Both rich hydrogen off gas stream and pure hydrogen stream are obtained at the outlet of the purification step.

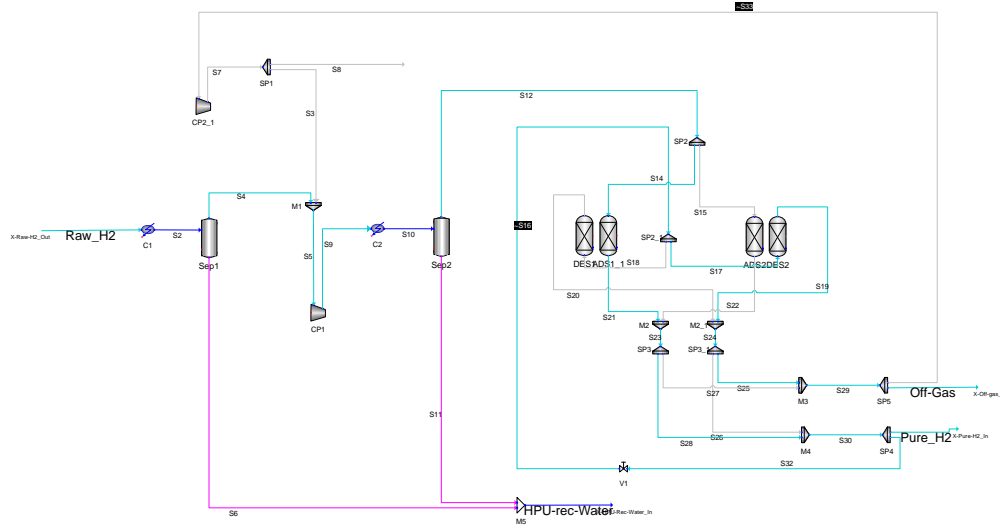


Figure 4. Symmetry® model of the HPU.

RESULTS AND DISCUSSIONS

The models once build, are used to simulate the DRI process for methane and hydrogen operation i.e., the conventional and the decarbonized DRI process. Methane is used as reducing agent and fuel for the conventional DRI process while electrolytic low-carbon hydrogen is considered for the decarbonized DRI process. The simulated performances of the DRI process are given in **Table I**.

Table I: Main output of both DRI process simulation for 50 t/h plant capacity

Feedstock		Exhaust Gas Recycled as Fuel		Electricity consumption (at the recycle compressor)	Direct CO ₂ emissions	Heat Availability (at the DRI gas exhaust)
Type	Flowrate (t/h)	Type	Flowrate (t/h)	kW	t/h	MW
Methane	6.2	34.7% CH ₄ 58.1% CO 6.8% CO ₂ 0.4 H ₂ O	6.8	6.1	4.3	21.1
Hydrogen	2.6	99.6% H ₂ 0.4% H ₂ O	0.7	3.9	0	21.7

35 t/h of direct reduced iron is obtained from 50 t/h of iron ore inlet stream. The model of the decarbonized scheme comprises the combination of the four models: (i) the DRI process, (ii) the heat recovery unit, (iii) the high temperature electrolyzer, and (iv) the hydrogen processing unit.

From the models, the global main reactant feedstock flowrates and fuel consumption are first estimated for the two schemes.

For the methane operation, it is observed higher recycle stream flowrate compared to hydrogen operation, leading to both higher fuel and electric consumption at the recycle compressor. In addition, the heat available at the DRI exhaust gas is found almost the same for both methane and hydrogen operation. Even if the flowrate of the DRI exhaust gas is more important for methane operation, its temperature is lower compared to hydrogen operation (539°C and 803°C for methane and hydrogen operation respectively). Note that the heat at the DRI exhaust gas is valorized for CO₂ capture purpose for methane operation.

In case of hydrogen operation, there is no CO₂ production. Indeed, no carbon-based compound feeds the process. The heat at the DRI exhaust gas is thus fully available and can be recovered as the electrolyzer thermal input. Accordingly, it is predicted a production of 2.4 t/h of electrolytic low-carbon hydrogen. An electrical input of 95 MWe is required at the electrolyzer. In such a use case, coupling the high temperature electrolyzer with the hydrogen DRI operation cannot fulfill the overall hydrogen demand of 2.6 t/h (as reducing agent and fuel), but still contribute to 92% of the hydrogen need.

CONCLUSIONS

In this study, methane replacement by electrolytic low-carbon hydrogen is assessed by mean of process modelling using the SLB proprietary computational thermodynamic process modelling software platform Symmetry®. Process models are thus built and calibrate to simulate performances of both conventional and decarbonized DRI process. The hydrogen operations are found more energy efficient than methane operations. In addition, the decarbonized scheme has no CO₂ direct emissions. With these considerations, HTE which is still a low maturity technology can be pointed out as a future-proof solution for integration with DRI process.

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