

# Hydrogen and Electricity Cogeneration System Based on Chemical Recuperation Enabled by Complementary Utilization of Natural Gas and Electricity

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## ABSTRACT

In this research, a hydrogen and electricity cogeneration system based on chemical recuperation enabled by complementary utilization of natural gas and electricity is proposed. The system uses an electrolysis for hydrogen generation and its byproduct oxygen for oxy-fuel burning to achieve near-zero CO<sub>2</sub> emissions. To increase the power generation, flue gas from turbine is used to reform with natural gas, decreasing irreversible losses in natural gas direct combustion. The thermodynamic analyses are conducted to analyze the performances of the proposed and reference systems. The results show that the energy and exergy efficiencies are 54.12% and 51.69%, respectively, improving by 2.96 and 2.93 percentage points compared to reference system.

**Keywords:** hydrogen and electricity cogeneration, chemical recuperation, pure oxygen combustion, near-zero CO<sub>2</sub> emission, thermodynamic analysis

## NONMENCLATURE

### Abbreviations

COM	Combustion Chamber
COMP	Compressor
CON	Condenser
EXC	Exchanger
REF	Reformer

### Symbols

$I$	Current density
$V$	potential
$\eta$	efficiency

## 1. INTRODUCTION

Currently, natural gas-fired power plants account for a quarter of the current world energy market[1]. Although its carbon emissions are substantially lower compared to coal-fired power plants, it still faces serious carbon emission problems and poses a large burden on the environment.

In order to solve carbon emissions, in addition to improving the thermodynamic efficiency of power plants[2], another method is to reduce CO<sub>2</sub> emissions by adding CO<sub>2</sub> capture devices[3]. Cheng et al.[4] proposed a retrofit design for existing natural gas combined cycle (NGCC) to achieve negative CO<sub>2</sub> emissions through the use of post-combustion carbon capture and direct air capture for power generation. To reduce the high cost and water consumption of carbon capture, De et al.[5] proposed a new low-cost carbon capture technology. This technique utilizes minimal energy to generate cold nitrogen chillers. A detailed thermodynamic modeling showed that a natural gas combined cycle power plant utilizing this technology can capture 99% of CO<sub>2</sub> with an energy of 631 MJ/tCO<sub>2</sub> and a water consumption of 574 L/tCO<sub>2</sub>. Moili et al.[6] used an aqueous solution of potassium tauroxylate instead of the conventional solvent of monoethanolamine for CO<sub>2</sub> capture in an NGCC power plant. This technique can reduce CO<sub>2</sub> emissions with low energy requirements. Nevertheless, capturing CO<sub>2</sub> requires additional energy and equipment, resulting in high energy consumption and high cost[7].

To achieve the goal of carbon neutrality, another method is to develop renewable energy sources. As the installed capacity of renewable energy generation gradually increases, the probability of the grid involving

the occurrence of supply-demand mismatch increases[8, 9]. Therefore, connecting an intermittent power source to an electrolyzer system to generate hydrogen is a good way to convert and store excess electrical energy[10]. Al-Buraiki et al.[11] electrolyzed water using an electrolyzer with the surplus power generated after satisfying the electrical load and the generated hydrogen was present in hydrogen storage tanks for use in vehicles. This scheme can produce more than 250 kg of hydrogen per year using the electrolyzer unit and the excess power was reduced from 64.9% to about 29%. Ali et al.[12] showed that the use of a 2kw electrolyzer in a system with a peak demand of 4kw can reduce the excess power

grade syngas before combustion, which reduces the irreversible loss of combustion and improves the power generation efficiency of the system; in addition, the system utilizes the discarded electricity through the electrolysis cell for the hydrogen production. At the same time, the system utilizes the by-product oxygen from the electrolysis cell to couple with the natural gas power generation system, replacing the traditional air separation unit to achieve low energy consumption and near-zero CO<sub>2</sub> emissions. It provides ideas for low-carbon and high-efficiency hydrogen and electricity co-generation.

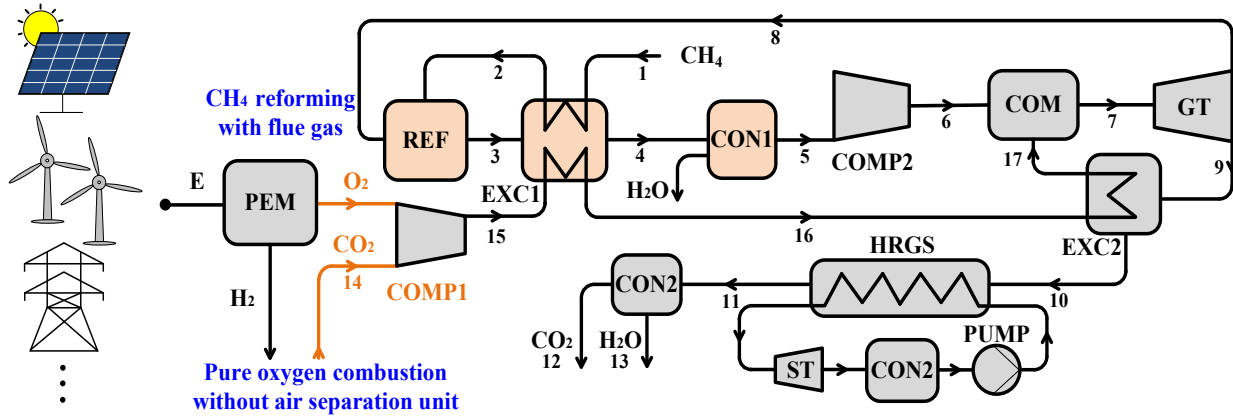


Fig. 1. Flowchart of the hydrogen generation system

in the system from 32.5% to 6%. Increasing the electrolyzer capacity to 3kw can reduce the excess power to less than 1%. In order to improve the efficiency of hydrogen production from the electrolyzer, many researchers have worked on the development of new catalysts and membrane materials[13], improvement of electrolyzer structure[14], and enhancement of external field technology[15]. The current researches on water electrolysis technology mainly focus on the hydrogen production process, and the utilization of oxygen (in situ consumption) is less discussed.

Although there are many studies on NGCC power plant with CCS and electrolysis of water, they still face many problems. Firstly, natural gas is directly combusted producing large irreversible losses; secondly, the extra devices for capture CO<sub>2</sub> results in a reduction of system efficiency; and finally, the by-product of hydrogen production by electrolysis of water is directly discharged into the air, which results in wastage.

To address the above problems, this paper proposes a hydrogen and electricity cogeneration system based on chemical recuperation of natural gas complemented with electricity. The proposed system utilizes the low-grade flue gas after gas turbine power generation to reform with high-grade natural gas to generate medium-

## 2. SYSTEM DESCRIPTION

### 2.1 Proposed system

Fig. 1 shows the flow diagram of the hydrogen and electricity cogeneration system with natural gas and electricity complementary. The exhaust gas from the gas turbine is divided into two parts: one part and natural gas enters the reformer and undergo a reforming reaction to generate high temperature syngas, and the heat from the high temperature syngas passes through a heat exchanger to preheat the natural gas and combustion gas. The cooled down syngas is cooled, compressed and then combusted with the by-product O<sub>2</sub> generated from the electrolysis cell in the combustion chamber for pure oxygen combustion, and the resulting high temperature flue gas is used in the GT to do work for power generation; the other part enters into the heat recovery steam generator after exothermic heating of the combustion gases through the heat exchanger for heat exchange, and then passes through the condenser for condensation to separate H<sub>2</sub>O and CO<sub>2</sub>. In order to avoid high temperatures during combustion of natural gas and oxygen in the COM, this can be controlled by regulating the CO<sub>2</sub> flow rate into the combustor.

## 2.2 Reference system

The reference system of this study (Fig. 2) consists of two parts: (a) an electrochemical hydrogen production system and (b) a gas-steam combined cycle power generation system using an air separation unit for oxyfuel combustion.

The PEM electrolysis uses excess electricity to electrolyze water into hydrogen and oxygen, producing hydrogen for subsequent use and oxygen as a by-product discharged directly into the air. In the power generation system, the natural gas is pressurized by a compressor and then enters the combustor, where it undergoes oxyfuel combustion with oxygen from an air separation unit, and the high-temperature flue gas enters the gas turbine. The flue gas from GT enters the HRGS to recover the sensible heat of the flue gas, which absorbs the sensible heat of the flue gas to produce steam and then goes to the steam turbine. The heat exchanged flue gas finally passes through the condenser for condensation to separate H<sub>2</sub>O and CO<sub>2</sub>. The reference system also controls the temperature of the combustion chamber by regulating the flow of CO<sub>2</sub> into the combustion chamber.

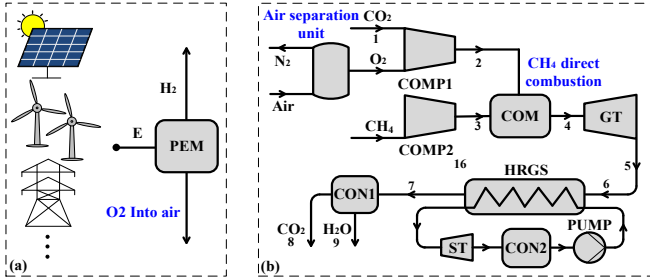


Fig. 2. Schematic diagram of the reference system.

## 3. MATERIAL AND METHODS

### 3.1 System simulation

The following assumptions were made during the simulation calculations: i) all processes were carried out in steady state; ii) the reactants were fully contacted in the reactor to reach chemical and phase equilibrium; and iii) the compression and transportation of separated CO<sub>2</sub> were not considered. The key parameters of the system during the simulation are shown in Table 1. Among them, the reformer adopts Gibbs reactor, with reforming temperature of 750 °C and reforming pressure of 1.01 bar; the combustion chamber adopts Stoic reactor, with combustion chamber outlet flue gas temperature of 1150 °C and pressure of 12 bar; the main steam temperature of the Rankine cycle is 565 °C and the pressure of 198 bar; the ambient temperature is 25 °C

and the pressure of 1.01 bar; and the electrolysis cell is based on PEM electrolysis cell as an example.

Table 1. Design parameters for the simulation.

Parameter	Value
Molar ratio of steam to carbon	2.5
Reaction temperature (°C)	750
Reaction pressure (bar)	1.01
Heat exchanger temperature difference(°C)	50
Pump isentropic efficiency (%)	88
Compressor isentropic efficiency (%)	88
Turbine isentropic efficiency (%)	92
Excess air coefficient	1.05
Combustion chamber heat loss (%)	0.05
Mechanical loss (%)	1
Pressure drops (%)	0

Exergy analysis can be used to determine the distribution of irreversible losses. At steady state, the equilibrium equations for the components in terms of exergy balance are expressed as follows:

$$\sum Ex_{in} - \sum Ex_{out} = \sum Ex_{loss} \quad (1)$$

For a complete equilibrium state, the exergy possessed by the system is the sum of the physical and chemical exergy, i.e., the expression:

$$\sum x = \sum x_{ph} + \sum x_{ch} \quad (2)$$

where the expression for physical exergy is as follows:

$$E_{x,ph} = m[(h - h_0) - T_0(s - s_0)] \quad (3)$$

where the expression for chemical exergy is as follows:

$$E_{x,ch} = -\Delta G + \sum_P n_j E_{x,m,j} - \sum_R n_j E_{x,m,j} \quad (4)$$

The electrolytic voltage of PEM is calculated as follows:

$$V = N_c \cdot (E_{ocv} + \eta_{act} + \eta_{ohm} + \eta_{diff}) \quad (5)$$

where  $N_c$  is the number of electrolytic cells;  $V_{ocv}$  is the open circuit voltage,  $V_{act}$  is the activation voltage;  $V_{ohm}$  is the ohmic voltage;  $V_{diff}$  is the diffusion voltage.

The formula for calculating the work consumed by the PEM electrolytic cell is as follows:

$$P = I \cdot V \quad (6)$$

where  $I$  is the current density of the electrolytic cell.

### 3.2 Evaluation

In this paper, the energy utilization rate and exergy efficiency are used as indicators to evaluate the

performance of the system. The energy utilization of the system is calculated as follows:

$$\eta_{th} = \frac{W_G + W_S - W_C + m_{H_2} LHV_{H_2}}{W_E + m_{CH_4} LHV_{CH_4}} \quad (7)$$

where  $W_G$  is the gas turbine power generation,  $W_S$  is the steam turbine power generation,  $W_C$  represents the total power consumption of all compressors and pumps, and  $W_E$  is the power consumption of the electrolysis cell, kW;  $LHV_{H_2}$  and  $LHV_{CH_4}$  are the lower heating value of hydrogen and methane, kJ/kg; and  $m_{H_2}$  and  $m_{CH_4}$  are the mass flow rates of hydrogen and methane, kg/s.

The exergetic efficiency of the system is as follows:

$$\eta_{ex} = \frac{E_G + E_S - E_C + E_{H_2}}{E_E + E_{CH_4}} \quad (8)$$

where  $E_G$  is the gas turbine output exergy,  $E_S$  is the turbine output exergy,  $E_C$  is the exergy consumed by all compressors and pumps,  $E_E$  is the input exergy from the electrolysis cell,  $E_{H_2}$  and  $E_{CH_4}$  are the hydrogen and methane output exergy, kJ/mol.

## 4. RESULTS AND DISCUSSION

### 4.1 Energy balance

Table 2 lists the thermodynamic characteristics of the main stream in the proposed system, together with its pressure, temperature, mass flow, and molar fraction. Table 2. Thermodynamic characteristics of the major streams in the proposed system.

Stream	Temperature (°C)	Pressure (bar)	Mole flow (mol/s)
1	25.00	1.01	1.00
2	265.60	1.01	1.00
3	750.00	1.01	5.46
4	210.00	1.01	5.46
5	25.00	1.01	4.93
6	317.46	12.00	4.93
7	1150.00	12.00	20.55
8	729.46	1.01	2.50
9	729.46	1.01	18.05
10	600.00	1.01	18.05
11	99.05	1.01	18.05
12	25.00	1.01	16.59
13	25.00	1.01	1.46
14	25.00	1.01	15.50
15	244.79	12.00	17.59
16	350.00	12.00	17.59
17	481.52	12.00	17.59

O2	25.00	1.01	2.09
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Through simulations, the energy balance between the hydrogen and electricity cogeneration system based on chemical recuperation natural gas complemented with electricity and the reference system is shown in Table 3. The results show that the energy inputs of the natural gas chemical and electrical energy of the two systems are 802.70 kW and 1443.11 kW, respectively. The hydrogen production of both systems is 1009.09 kW. The net electricity production of the proposed system is 418.23 kW, which is increased by 66.64 kW compared to the 351.58 kW of the reference system. The energy efficiency of the proposed system is 63.55%, which is 2.96 percentage points higher than that of the reference system of 60.59%.

Table 3. Energy balance between the proposed and reference systems.

Item	Proposed system		Reference system	
	Value (kW)	(%)	Value (kW)	(%)
<b>Energy input</b>	<b>2245.81</b>	<b>100.00</b>	<b>2245.81</b>	<b>109.79</b>
Natural gas	802.70	35.74	802.70	35.74
Electricity	1443.11	64.26	1443.11	64.26
<b>Self-consume</b>	<b>202.09</b>	<b>9.00</b>	<b>219.81</b>	<b>9.79</b>
Compressor and pump	202.09	9.00	165.62	7.37
Air separation	-	-	54.19	2.41
<b>Energy output</b>	<b>1427.31</b>	<b>63.55</b>	<b>1360.67</b>	<b>60.59</b>
H <sub>2</sub>	1009.09	44.93	1009.09	44.93
Electricity	418.23	18.62	351.58	15.66
<b>Energy losses</b>	<b>818.50</b>	<b>36.45</b>	<b>885.15</b>	<b>39.41</b>
Heat transfer	21.69	0.97	0.00	0.00
Condensation	50.94	2.27	0.00	0.00
HRGS	272.92	12.15	347.92	15.49
Flue gas	26.13	1.16	43.62	1.94
PEM	434.03	19.33	434.03	19.33
Air separation	-	-	54.19	2.41
Others	12.78	0.57	5.38	0.24
<b>Energy efficiency, %</b>	<b>63.55</b>		<b>60.59</b>	

It can be seen from Fig. 3 that in both systems, PEM losses account for the largest proportion of all losses, which is 19.33%. The HRGS loss of the proposed system is 12.15%, which is 3.34 percentage points lower than 15.49% of the reference system. In the reference system, due to the use of air separation device to produce oxygen, the energy consumption of this component accounts for 2.41%. Because the proposed system uses oxygen, a byproduct of PEM electrolysis of water, there is no such loss.

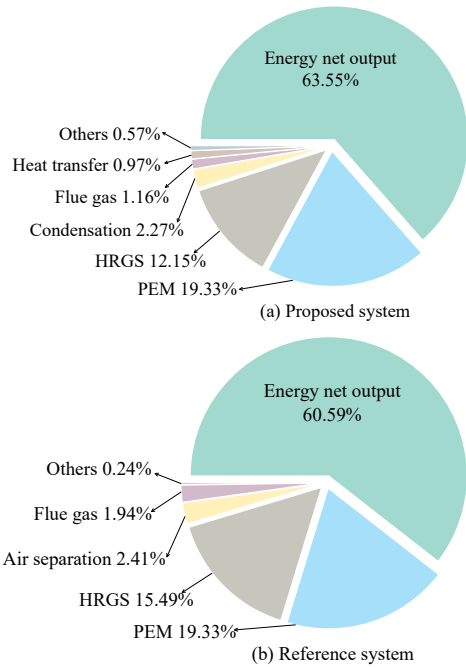


Fig. 3. Energy balances of the proposed and reference systems.

#### 4.2 Exergy balance

In order to further reveal the mechanism of the improved performance of the proposed system, exergetic balance analysis of the proposed system and reference system has been carried out, and the analysis results are shown in Table 4. When both systems have the same amount of fuel and power inputs, the proposed system has an exergy efficiency of 61.48% and the reference system has an exergy efficiency of 58.55%, which is an improvement of 2.93 percentage points in the exergy efficiency of the proposed system over the reference system

Table 4. Exergy balances of the proposed and reference systems.

Item	Proposed system		Reference system	
	Value (kW)	(%)	Value (kW)	(%)
<b>Exergy input</b>	<b>2277.51</b>	<b>100.00</b>	<b>2277.51</b>	<b>100.00</b>
Natural gas	834.40	36.64	834.40	36.64
Electricity	1443.11	63.36	1443.11	63.36
<b>Self-consume</b>	<b>202.09</b>	<b>8.87</b>	<b>219.81</b>	<b>9.65</b>
Compressor and pump	202.09	8.87	165.62	7.27
Air separation	-	-	54.19	2.38
<b>Exergy output</b>	<b>1400.20</b>	<b>61.48</b>	<b>1333.56</b>	<b>58.55</b>
H <sub>2</sub>	981.98	43.12	981.98	43.12
Electricity	418.23	18.36	351.58	15.44
<b>Exergy losses</b>	<b>877.31</b>	<b>38.52</b>	<b>943.95</b>	<b>41.45</b>
Reformer	28.16	1.24	0.00	0.00

Combustor	194.91	8.56	260.68	11.45
Heat transfer	39.16	1.72	0.00	0.00
HRGS	63.40	2.78	100.22	4.40
Condensation	8.06	0.35		0.00
Flue gas	23.59	1.04	5.73	0.25
Air separation	-	-	54.19	2.38
Others	39.03	1.71	41.97	1.84
PEM	461.14	20.25	461.14	20.25
CO <sub>2</sub> output	19.86	0.87	20.03	0.88
<b>Exergy efficiency, %</b>	<b>61.48</b>		<b>58.55</b>	

As can be seen in Fig. 4, the combustion losses are the second largest for both the two systems, in addition to the largest losses for the PEM. This is different from Fig. 3 where the second largest loss is the HRSG loss. The combustion loss of the proposed system is 8.56%, which is 2.89 percentage points lower compared to the 11.45% of the reference system. This is due to the reduction of irreversible losses in the combustion process after the reforming of high-grade methane with a portion of low-grade high-temperature flue gas to produce medium-grade syngas as compared to direct combustion of methane. The process achieves a cascade utilization of different grades of energy. The air separation unit in the reference system has 2.38% exergy loss. The proposed system has additional reformer and heat exchanger due to increased reforming process and better utilization of waste heat, which increases the exergy loss by 1.24% and 1.72% respectively.

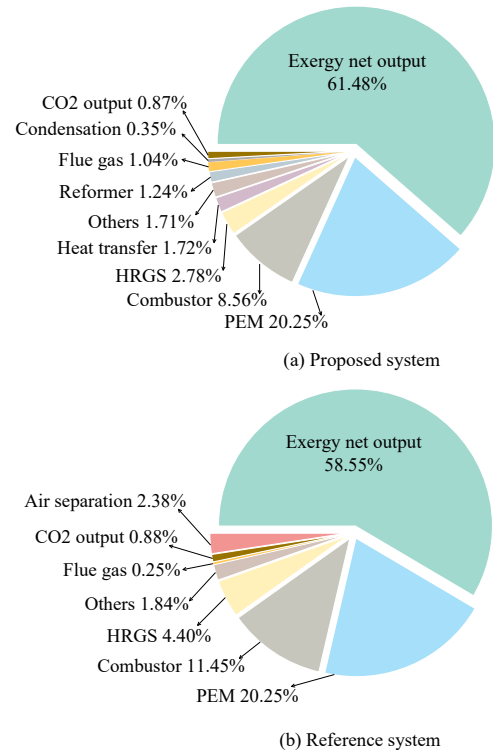


Fig. 4. Exergy balances of the proposed and reference systems.

## 5. CONCLUSION

In this study, a combined hydrogen and power generation system based on complementary natural gas and electricity with chemical recuperation is proposed. The thermodynamic performances of the proposed system and the reference system are carried out, and the following conclusions have been discovered:

(1) The energy and exergy efficiencies of the proposed system are 63.55% and 61.48%, respectively, which are 2.96 and 2.93 percentage points higher compared to the reference system with near-zero CO<sub>2</sub> emissions.

(2) The system eliminates the energy-consuming air separation device, which saves 54.19 kW and improves the energy efficiency by 2.41% compared to the reference system.

(3) Another key process for the performance improvement of the proposed system is the reduction of irreversible losses in the combustion process by 65.77 kW and the efficiency improvement of 2.89% due to the use of chemical recuperation.

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## DECLARATION OF INTEREST STATEMENT

We declare that we have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. All authors read and approved the final manuscript.

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