Integrated methanol-hydrogen-electricity conversion system with low CO₂ emission for on-demand power and heat supply[#]

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ABSTRACT

To address climate change, distributed energy system shall explore a synergistic pathway towards high efficiency and low carbon emissions while ensuring the continuity, stability and variety of energy supply. We propose a novel distributed energy system featuring the integration of methanol steam reforming, metal alloybased hydrogen separation, proton exchange membrane fuel cells and CO₂ capture. Methanol, as a convenient, stable, and economical medium for hydrogen storage and on-demand power generation, serves as a transition fuel between current and future energy systems. Using the toxicity-resistant LaNi_{4.3}Al_{0.7}, hydrogen is absorbed from methanol reformate gas containing CO and water vapor at 180°C and then released through isothermal depressurization. Simultaneously, the tail gas from the hydrogen separation unit undergoes oxy-fuel combustion, achieving CO₂ capture with low energy consumption. Subsequently, the proton exchange membrane fuel cells efficiently convert hydrogen to electricity, with the waste heat being exchanged to supply hot water. The models of reformer, metal alloybased hydrogen separation unit and proton exchange membrane fuel cells are validated by experimental data from the literature. The analysis demonstrates that at 100 mol h⁻¹ methanol flow rate, 250°C reaction temperature and 8 bar absorption pressure, total electrical efficiency and total energy efficiency of the proposed system are 40.05% and 88.00%, respectively, which are 5.99 and 1.2 percentage points higher than those of the reference system with conventional pressure swing adsorption method. Meanwhile, the total exergy efficiency of the proposed system is 43.38%, which is 5.5 percentage points higher than the reference system. The system enables an efficient on-demand supply of power and heat, the potential for reducing CO_2 emission, and compatibility with future energy systems.

Keywords: distributed energy system, methanol reforming, hydrogen, metal alloy, CO₂ capture

NONMENCLATURE

Abbreviat	tions
PSA	Pressure Swing Adsorption
PEMFC	Proton Exchange Membrane Fuel Cell
НΧ	Heat Exchanger
GT	Gas Turbine
Symbols	
Q_{sloar}	Input Solar Energy
η_{abs}	Absorption Efficiency of Solar Collector
$\eta_{ m opt}$	Optical Efficiency of Solar Concentrator
Qin	Thermal Energy Input into PEMFC
Qout	Thermal Energy Output from PEMFC
$Q_{\rm cw,fc}$	Waste Heat from PEMFC
WPEMFC	Power Output of PEMFC
W _{tot}	Net power output of system
W _{GT}	Power Generated by GT
W _{ASU}	Power Consumption of Air Separation Unit
W _{COM}	Power Consumption of Compressors
W_{pump}	Power Consumption of Pumps
n _{me}	Molar Flow Rate of Methanol
HHV_{me}	Higher Heating Value of Methanol
Q_{DHW}	Heat Provided by Domestic Hot Water
EX heat	Exergy Flow Output of Heat
EX solar	Solar Exergy Flow
<i>EX</i> _{me}	Exergy Flow Input of Methanol

1. INTRODUCTION

To effectively address global climate change, the development and deployment of low-carbon energy technologies are imperative, aiming to reduce global greenhouse gas emissions to 25 GtCO_{2-eq} yr⁻¹ by 2030^[1]. In particular, residential and commercial energy consumption within the 1 kW to 1 MW range accounts

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for approximately 30-40% of global electricity consumption^[2], with considerable potential for carbon reduction. Small-scale distributed energy systems suitable for residential and commercial application require a transition from the traditional chain development pattern to the synergy development pattern to balance energy efficiency and carbon reduction.

Hydrogen, with its high energy density and cleanliness, stands out as an excellent energy carrier for small-scale distributed energy systems ^[3, 4]. However, due to its low bulk density, flammability and explosiveness, hydrogen transportation and storage are challenging^[5], constituting a bottleneck for the development of distributed energy systems involving fuel cells. Methanol is liquid at ambient temperature and pressure that facilitates the utilization of existing energy transportation and storage infrastructures, rendering it a convenient, stable, and economical hydrogen storage medium for distributed energy systems^[6]. Moreover, methanol can be synthesized from fossil fuels or renewable sources, serving as a transition fuel between current and future energy systems. Furthermore, methanol steam reforming for hydrogen production can be driven by renewable energy sources such as solar thermal energy^[7] and geothermal energy^[8], achieving energy efficiencies of approximately 60-80%.

Combining hydrogen production with cogeneration systems presents benefits in energy utilization.

Nevertheless, separating high-purity hydrogen from methanol steam reforming gas is critical due to byproducts. Separation methods include pressure swing adsorption (PSA), selective membrane separation and metal alloy separation^[9-11]. Compared with other methods, hydrogen separation method based on rareearth metal alloys exhibit beneficial features such as high hydrogen purity, hydrogen recovery rate and reliability. For instance, the AB5-type metal alloy system can achieve a 94% hydrogen recovery rate while maintaining purity between 99-99.9%^[12]. Additionally, research indicates that the LaNi5-type alloys have the ability to absorb and separate hydrogen even in challenging environments, as demonstrated by the fluorinated LaNi_{4.3}Al_{0.7} alloy prepared by Hao et al.^[13] which maintains hydrogen absorption capacity in a gas mixture containing toxic gases at elevated temperatures.

Tail gas from hydrogen separation unit is generally combusted to supply heat, followed by CO₂ capture. However, the low CO₂ concentration in the flue gas necessitates high energy consumption for CO₂ capture. Oxygen-enriched combustion simplifies CO₂ separation by utilizing high-purity oxygen (purity>95%), as demonstrated by Wu et al.^[14], where solid oxide fuel cell anode exhaust gas is oxygen-enriched combusted to generate high-temperature flue gas primarily comprising CO₂ and water vapor, facilitating CO₂ concentration enrichment with minimal energy consumption through water vapor condensation.



Fig. 1 Diagram of the methanol-hydrogen-electricity conversion distributed energy system integrating metal alloy separation unit with CO₂ capture.

Based on the literature review, we propose a novel distributed energy system integrating methanol steam reforming, metal alloy-based hydrogen separation, proton exchange membrane fuel cell (PEMFC) and CO₂ capture. An integral model of the proposed system is developed and model validations are carried out for the reformer, the hydrogen separation unit and the PEMFC. A reference system integrating the conventional PSA is established for comparison. Energy and exergy analyses are conducted to assess system performances. The proposed system, with its dual attributes of high efficiency and low carbon emissions, holds significance for decarbonizing small-scale distributed energy systems.

2. SYSTEM DESCRIPTION

2.1 Proposed system

Fig.1 is a diagram of the proposed methanolhydrogen-electricity conversion distributed energy system, including a solar thermochemical subsystem, a gas processing subsystem and a PEMFC subsystem.

In the solar thermochemical subsystem, methanol is pumped from a storage tank into the heat exchanger (HX)-1, the solar-driven preheater, and ultimately into the solar-driven reformer. Another reactant, water, is pumped from its storage tank into HX-2, where it is vaporized and heated to the reformer temperature driven by the high temperature flue gases coming from the gas turbine (GT) and then enters the reformer. In the reformer, most reactants are converted to syngas through the methanol steam reforming. The syngas then passes through HX-1 for heat recovery and enters the gas processing subsystem.

In the gas processing subsystem, syngas at 180° C enters the hydrogen separation unit containing the metal alloy to obtain high purity hydrogen (CO<20 ppm), which is then sent to the PEMFC subsystem. The tail gas undergoes oxy-fuel combustion, producing high-temperature flus gas with CO₂ and water vapor. The flue gas passes through the GT for power generation and heat recovery. After condensation and water separation, high-purity CO₂ is obtained, enabling effective CO₂ capture.

In the PEMFC subsystem, high-purity hydrogen is supplied as the anode fuel, while preheated air is fed into the cathode as the oxidizer. While generating electricity, the PEMFC also produces a significant amount of heat. Water is used as a coolant to maintain the PEMFC at its designed operating temperature. Additionally, the coolant exchanges heat with cold water to provide domestic hot water.

2.2 Reference system

A reference distributed energy system employing conventional PSA for hydrogen separation is presented for comparison with the proposed system, as shown in Fig. 2. The primary distinction between the two systems lies in the hydrogen separation unit within the red cycle in Fig. 2. According to the literature, the hydrogen recovery rate of the separation unit in the reference system is set to 73.3% under similar operating pressure as the proposed system. We assume that the operating parameters of the remaining components in the reference system align with those of the proposed



Fig. 2 Diagram of the reference distributed energy system integrating conventional pressure swing adsorption hydrogen separation unit.

system.

3. METHODOLOGY

3.1 System simulation

The system is simulated using Aspen Plus software, with the Peng-Robinson method selected for calculating physical properties. The main assumptions of the simulation are as follows:

- The system operates under steady-state conditions, with pressure losses in pipes and components disregarded.
- (2) Heat exchange losses between the system and the external environment, as well as losses among various heat exchangers, are neglected.
- (3) Air is considered an ideal gas, composed of 79% N_2 and 21% O_2 by volume.
- (4) The hydrogen delivered to the PEMFC is assumed to be of 100% purity, with the fuel cell operating at uniform internal temperature and pressure, and the

operating conditions of the PEMFC stack consistent with those of a single fuel cell.

3.1.1 Solar thermochemical subsystem

The reformer is established with the plug flow reactor model in Aspen Plus, utilizing the widely recognized CuO/ZnO/Al₂O₃ catalyst and considering the three primary reactions: methanol steam reforming, methanol decomposition and water-gas shift. The kinetic model proposed by Peppley et al.^[15] is employed for the reformer.

The required input solar energy for the preheater and reformer is shown below:

$$Q_{\rm solar} = \frac{Q_{\rm heat}}{\eta_{\rm abs} \eta_{\rm opt}} \tag{1}$$

where Q_{heat} is the heat required for preheating and reaction, η_{abs} is the absorption efficiency of solar collector, η_{opt} is the optical efficiency of the solar concentrator.

3.1.2 Gas processing subsystem

The cycle model of the hydrogen separation unit is established through an external program, with each cycle comprising four steps: pressurization, absorption, depressurization, and desorption. Hydrogen with a purity greater than 99% and CO content below 20 ppm is considered usable, and the hydrogen recovery rate is determined under the system's operating condition. Furthermore, in the whole system model, a separator represents the hydrogen separation unit, with the separation ratio (i.e., hydrogen recovery rate) indicating the hydrogen separation performance.

To minimize fuel wastage and facilitate CO₂ capture, the tail gas from the hydrogen separation unit is fed into the micro-GT for oxy-fuel combustion and power generation. The SGT-300 GT with high energy utilization efficiency and low cost is used in this study.

3.1.3 PEMFC subsystem

Due to the absence of a built-in model for fuel cells in Aspen Plus, external programming is employed to build the electrochemical model and energy balance of the PEMFC. The electrochemical model of PEMFC presents the relationship between cell voltage and current density, considering activation loss, ohmic loss, and concentration loss, as detailed in reference^[16]. The heat generated by the PEMFC is primarily carried away by the outlet gas and cooling water, and the energy conservation of PEMFC is expressed by the following equation:

$$Q_{\rm in} = Q_{\rm out} + Q_{\rm cw,fc} + W_{\rm PEMFC}$$
(2)

where Q_{in} is the energy input into the PEMFC, Q_{out} represents the energy output from the PEMFC, and $Q_{cw,fc}$ is the waste heat from the PEMFC.

3.2 Evaluation metrics

The net electrical generation of the system is:

$$W_{\text{tot}} = W_{\text{out}} + W_{\text{GT}} - W_{\text{ASU}} - W_{\text{COM}} - W_{\text{pump}}$$
(3)

where W_{GT} is the power generated by GT; W_{ASU} , W_{COM} and W_{pump} represent the power consumption of the air separation unit, compressors, and pumps, respectively. The total electrical efficiency is obtained as:

$$\eta_{\rm e} = \frac{W_{\rm tot}}{n_{\rm me} \rm HHV_{\rm me} + Q_{\rm solar}} \tag{4}$$

where n_{me} is the molar flow rate of methanol, and HHV_{me} represents the higher heating value of methanol.

Based on the first law of thermodynamics, the total energy efficiency is calculated as follows:

$$\eta_{\rm en} = \frac{W_{\rm tot} + Q_{\rm DHW}}{n_{\rm me} \rm HHV_{\rm me} + Q_{\rm solar}}$$
(5)

where Q_{DHW} is the amount of heat contained in the hot water supplied.

Based on the equations provided in literature ^[14], the exergy of the mixed stream, the exergy balance of the components, and the solar exergy flow are calculated. The total exergy efficiency is defined as:

$$\eta_{\rm en} = \frac{W_{\rm tot} + EX_{\rm heat}}{EX_{\rm solar} + EX_{\rm me}}$$
(6)

where EX_{heat} is exergy flow output of heat, EX_{solar} is solar exergy flow, and EX_{me} is exergy flow input of methanol.

4. RESULTS AND DISCUSSION

4.1 Model validation and operating conditions

The models of the reformer, the metal alloy-based separation unit, and the PEMFC are validated, as shown in Fig. 3. In the reformer, the maximum relative error between the simulated and experimental methanol conversion rates is 3.42% at temperatures ranging from 513K to 533K. For the metal alloy's absorption and desorption performance at 180° C, with an atmosphere of 99%H₂ and 1%CO, 0.9 MPa for absorption, and 0.5 MPa for desorption, the simulation results closely match the experimental results. For the electrochemical model of the PEMFC, at 348K and 1 atm, the maximum relative error between the simulated power output and the experimental results is 4.83%. These relative errors are within an acceptable range, ensuring the reliability of the model for subsequent calculations.

The operating conditions and key parameters are summarized in Table 1. The methanol input flow rate,



Fig. 3 The validation between experimental results and simulation results for (a) reformer^[15], (b) Absorption and desorption performance of metal alloy^[13], and (c) PEMFC^[16].

operating parameters of reformer, burner, GT and PEMFC are identical for the proposed system and the reference system. However, the hydrogen separation unit of the reference system operates at 40°C, which is different from the proposed system. Moreover, the pressure of the output high-purity hydrogen is 8 bar and the pressure of the tail gas is 1 bar.

Table 1 Basic operating conditions and key parameters

Item	Value
Reformer temperature (°C)	250
Reformer pressure (bar)	8
Methanol flow rate (mol h ⁻¹)	100
Molar ratio of water to methanol	1
Mechanical efficiency of the pump	0.9
Minimum temperature difference in the heat	
exchangers (°C)	15
Optical efficiency	0.87
Absorption efficiency of solar collector	0.9
Isentropic efficiency of the gas turbine	0.85
Mechanical efficiency of the gas turbine	0.98
Inlet temperature of the gas turbine (°C)	945
Isentropic efficiency of the compressor	0.82
Mechanical efficiency of the compressor	0.99
Cell operation temperature (°C)	85
Cell operation pressure (bar)	1
Number of single fuel cells	90
Active area (cm ²)	200
Oxygen stoichiometry	1.15
DC/AC conversion of efficiency	0.92
Inlet temperature of cold water (°C)	25
Temperature of hot water (°C)	55

4.2 Results of hydrogen separation unit

Firstly, the simulation results for the hydrogen separation unit are presented. The pressurization and absorption steps are fed with 391.3 mol h⁻¹ syngas from the reformer. The outlet pressure is 8 bar and the desorption pressure is 1 bar. The time and average outlet components for each step are detailed in Table 2.

Hydrogen with a CO concentration below 20ppm is considered usable, as indicated by the red-filled portions in Fig. 4 (b). Simulation outcomes reveal an average hydrogen purity of 99.87%, with a recovery rate of 92.77%. The main impurity in hydrogen, CO_2 , averages 0.12%, demonstrating a negligible impact on the performance of the PEMFC.

Table 2	Average	outlet	com	onents	for	each	step
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Char	Time	Outlet
Step	(s)	CH ₃ OH/H ₂ O/H ₂ /CO ₂ /CO (%)
Pressurization	20	-
Absorption	440	3.63/4.04/14.07/77.85/0.41
Depressurization	20	0.62/0.69/85.35/13.27/0.07
Desorption	440	0.006/0.006/99.87/0.118/0.0006

4.3 Energy analysis

The overall energy conversion performances of the proposed system and the reference system are presented in this section. Table 2 presents the energy performances of the two systems at a methanol molar rate of 100 mol h⁻¹. The solar energy input of the proposed system is 16.22 percentage points higher than that of the reference system, with the main difference being the amount required to preheat methanol. In the proposed system, methanol is preheated in HX-2 using the sensible heat of the syngas from the reforming temperature (250°C) to the hydrogen separation unit temperature (180°C), and then further heated by solar energy input. While in the reference system, where the syngas is reduced from 250°C to 50°C in the HX-2 for preheating the reactant methanol, the extra input of solar energy for preheating is relatively lower.

With the hydrogen recovery rate elevated from 73.3% to 92.77%, the proposed system demonstrates enhanced energy conversion performance. The net electric power output stands at 9.43 kW, surpassing the reference system by 1.6 kW. Correspondingly, the proposed system achieves total electrical efficiency and total energy efficiency of 40.05% and 88.00%,

respectively, exceeding the reference system by 5.99 percentage points and 1.2 percentage points.

Table 3 Energy performances of the proposed system and reference system

Item	Proposed	Reference
	system	system
Total energy input rate (kW)	23.55	23.00
Methanol energy input rate (kW)	20.16	20.16
Solar energy input rate (kW)	3.39	2.84
Net energy output rate (kW)	20.72	19.96
Power output from PEMFC (kW)	10.03	8.16
Power output from GT (kW)	0.95	2.61
Overall heat output rate (kW)	11.30	12.13
Net electric power output (kW)	9.43	7.83
Total electrical efficiency	40.05	34.06
Total energy efficiency	88.00	86.8

Fig.5 illustrates the detailed energy output distributions of the two systems. In Fig. 5 (a) and Fig. 5 (b), the total power generation from PEMFC and GT are used as the base value (100%). In the proposed system, power generation of PEMFC is 10.02 kW, of which 0.8 kW is lost for DC/AC conversion, resulting in a net output of 9.22 kW. The electricity generated by GT serves to power





system components, with COM-2 being the largest power consumer at 0.2 kW. The remaining power output of GT is 1.88%. In the reference system, the power generation of the PEMFC is lower at 8.16 kW due to the lower hydrogen recovery rate. Correspondingly, the power generation of GT is larger in the reference system. However, despite increased oxygen and CO₂ circulating for combustion, GT output only slightly improves at 0.32 kW. Furthermore, illustrated in Fig. 5(c), the reference system supplies an additional 0.83 kW of hot water, attributed to the combustion of a larger quantity of hydrogen.

4.4 Exergy analysis

Exergy analyses are carried out to compare the work capacity of the two systems, as presented in Table 4. The total exergy flow input of the proposed system is 24.12 kW, of which methanol contributes 20.96 kW and solar energy contributes 3.16 kW. The total exergy flow output is 10.46 kW with a total exergy efficiency of 43.38%, of which the electrical and thermal outputs are 9.43 kW and 1.03 kW, respectively. In comparison, the reference system has an exergy efficiency of 37.88%, which is 5.5 percentage points lower than the proposed system.

Table 4 Exergy performances of the proposed system and reference system

Itom	Proposed	Reference	
Item	system	system	
Total exergy flow input (kW)	24.12	23.61	
Exergy flow input of CH₃OH (kW)	20.96	20.96	
Exergy flow input of sunlight (kW)	3.16	2.65	
Total exergy flow output (kW)	10.46	8.94	
Exergy flow output (electricity) (kW)	9.43	7.83	
Exergy flow output (thermal) (kW)	1.03	1.11	
Total exergy loss rate (kW)	13.08	14.67	
Total exergy efficiency (%)	43.38	37.88	

5. CONCLUSIONS

A novel distributed energy system has been proposed, which enables flexible utilization of solar energy, facilitates dispatchable cogeneration, and holds potential for decarbonization. High hydrogen purity (99.87% (CO <20 ppm)) and high hydrogen recovery rate (92.77%) are achieved by the metal alloy-based hydrogen separation unit. The proposed system is reference compared with the system where conventional pressure swing adsorption method is employed as the hydrogen separation unit. Thermodynamic analysis indicates that the proposed system is to be more efficient than the reference system. The total electrical efficiency and total energy efficiency of the proposed system are 40.05% and 88.00%,

respectively, 5.99 percentage points and 1.20 percentage points higher than the reference system. Meanwhile, the proposed system is more competitive in terms of work capacity than the reference system. The total exergy efficiency of the proposed system is 43.38%, which is 5.5 percentage points higher than the reference system.

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