

High Efficiency SNG Production System based on High Temperature Electrolysis and Biomass Gasification

Dandan Wang^{1*}, Yalou Li¹, Yuze Dai^{2,3}

1 China Electric Power Research Institute, Beijing 100192, PR China

2 University of Chinese Academy of Sciences, Beijing 100049, PR China

3 Institute of Engineering Thermophysics, Chinese Academy of Sciences, Beijing 100190, PR China

ABSTRACT

The intermittence and site-dependence of renewables requires the efficient systems to storage the surplus wind/solar electricity. Substitute nature gas (SNG) is a promising energy carrier which facilitates existing pipeline transportation. This work proposed a novel efficient SNG production system, in which the high temperature electrolysis combined with oxygen/steam biomass gasification is adopted. In this proposed system, the O₂ produced by electrolyzer is sent to the gasifier directly and the air separation can be eliminated. Besides, through the coexistence methanation process, the conventional water gas shift process and CO₂ capture consumption will be avoided. The electrochemical model of SOEC is established and the whole SNG production system is simulated by Aspen plus. Parametric and energy/exergy analyses are conducted and the results shown that the thermal efficiency of the novel plant can reach 77.4%, which is 15 percentage points higher than the traditional SNG production pathways. This work provide a new method to realize high energy conversion efficiency and reduce the wind/solar energy curtailment.

Keywords: SNG production, high temperature electrolysis, steam/oxygen biomass gasification, thermodynamic analyses

1. INTRODUCTION

Renewable energy sources, such as wind, solar, hydropower and biomass are considered as the promising alternatives to the traditional fossil fuels [1,2]. Among these, the wind and solar power develop rapidly

in recent years, while the character of intermittence and site-dependence causes the uncontrollable production and reduces the commerciality of these sources. In 2018, China's renewable energy surplus reached 102 billion kWh, exceeding the annual generation of the world biggest hydropower station, the Three Gorges Dam [3].

Energy storage provide a feasible pathway to reduce the mismatch between the power production and demand of these intermittent resources. Power-to-gas (P2G) refers one vital chemical energy storage technology which transforms the surplus renewable energy to gaseous chemicals such as hydrogen or substitute nature gas (SNG) [4,5]. Compared with H₂, SNG facilitates existing pipeline transportation and is more convenient for long-term storage through shifts in capacity between energy networks [6].

Current Power-to-Gas technology has an energy conversion efficiency around 55%-60%, including the water electrolysis and CO₂ Sabatier process [7]. Moreover, the required carbon source usually derives from the flue gas or biogas or atmosphere with extra energy consumption for CO₂ separation. Therefore, how to improve the energy efficiency and avoid large consumption on the carbon source acquisition is significant for the commercial operation of P2G plants.

This paper proposed a high efficiency SNG production system: SOEC relied on the renewable power produces hydrogen as the energy carrier, and provides O₂ as gasification agent for gasifier to improve the thermal efficiency of biomass; the renewable hydrogen combined with syngas from biomass gasification will be used for CO₂/CO coexistence methanation process,

which can break through the conventional idea of water gas shift process and avoid CO₂ separation consumption.

In this way, the effective utilization of resources and heat energy can be realized, which will significantly improve the energy efficiency of the whole power-to-gas system. Since biomass is the only renewable carbon source, the coupling process achieves zero carbon emission in the full life cycle.

2. DESIGN AND MODELING OF THE SNG PLANT

2.1 Description of system configuration

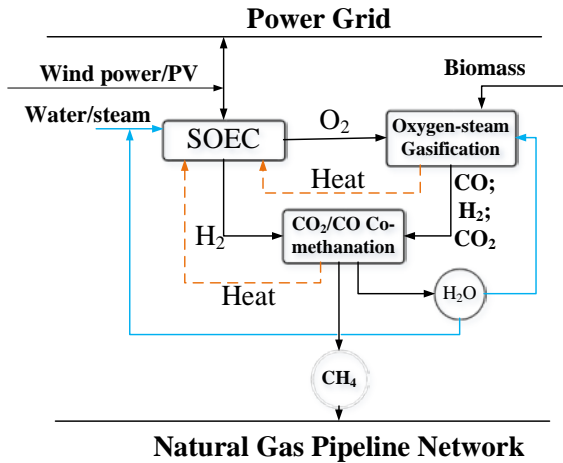


Fig. 2 Simple diagram of the proposed SNG plant

The proposed SNG plant mainly contains the high-temperature solid oxide electrolysis unit, oxygen/steam biomass gasification unit and CO₂/CO co-methanation unit and its simple diagram is shown in Fig. 1 [8][9].

The SOEC is driven by the renewable electricity and the process heat of waste heat boiler and methanation process. The steam is preheated by anode products, and then enters the cathode side of the SOEC. The anode O₂ product is used for gasification process, and the H₂ is pressurized and further sent to co-methanation process. Steam from the methanation process and O₂ from SOEC are used for gasifying agents. After purification, the clean syngas will mix with the hydrogen from SOEC and the H₂/(CO+CO₂) is set to be 3. The mix fuel gas reacts in the co-methanation unit to obtain the final CH₄ for pipeline transportation.

Using the by-products and heat recovery appropriately has positive impact on the whole plant efficiency and could be cost effective.

2.2 Modeling of Solid Oxide Electrolyzer Cell

The high temperature electrolysis is the key process of the proposed SNG plant and its detailed

electrochemical model is established by MATLAB [10,11].

The SOEC is composed of the porous cathode electrode and anode electrode on both sides and a dense electrolyte layer in the middle. Steam are sent into the cathode to generate hydrogen and oxygen ions, and 10% hydrogen comes with steam to prevent oxidation of the cathode in the high temperature and high humidity environment. The cations are transferred to the anode

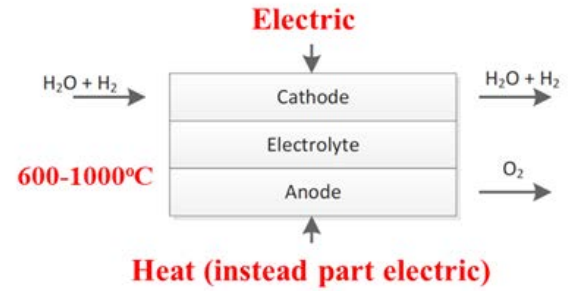
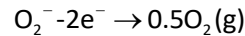
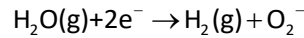


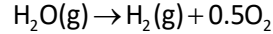
Fig. 1 High Temperature Solid Oxide Electrolysis

side through the electrolyte and converted into oxygen, as shown in Fig. 2.

The cathodic reaction and the anodic reaction is as follows:



The total reaction of the SOEC can be expressed as:



In the process of electrolysis, the practical electrode potential is higher than the equilibrium potential due to the irreversible destruction, which is called polarization phenomenon. This causes part of electric energy converted into heat energy. The polarization losses includes concentration polarization E_{con} , activation polarization E_{act} and ohmic polarization E_{ohm} . The actual cell potential E can be obtained:

$$E = E_r + E_{con} + E_{act} + E_{ohm}$$

Where E_r is the equilibrium potential.

$$E_r(T, P) = \frac{\Delta G}{nF} + \frac{RT}{nF} \ln\left(\frac{P_{\text{H}_2} P_{\text{O}_2}^{0.5}}{P_{\text{H}_2\text{O}}}\right)$$

Concentration overvoltage E_{con} caused by diffusion includes cathode and anode concentration overvoltage:

$$E_{con,c}^{\text{SOEC}} = \frac{RT}{nF} \ln\left(\frac{P_{\text{H}_2}^1 P_{\text{H}_2\text{O}}^0}{P_{\text{H}_2}^0 P_{\text{H}_2\text{O}}^1}\right)$$

$$E_{con,a}^{\text{SOEC}} = \frac{RT}{nF} \ln\left[\left(\frac{P_{\text{O}_2}^1}{P_{\text{O}_2}^0}\right)^{0.5}\right]$$

The activation overvoltage is calculated by Butler-Volmer formula:

$$E_{act}^{SOEC} = \frac{RT}{F} \ln \left[\frac{J}{2J_{0,i}} + \sqrt{\left(\frac{J}{2J_{0,i}}\right)^2 + 1} \right]$$

The activation overvoltage is as follows:

$$E_{ohm}^{SOEC} = 2.99 \times 10^{-5} J d_E \exp\left(\frac{10300}{T}\right)$$

The entropy generation S_{gen} of the electrolysis process is related with the over polarizations, the number of electron transferred n and the temperature T , and S_{gen} is as follows:

$$S_{gen} = \frac{nF(E_{ohm} + E_{con} + E_{act})}{T}$$

F is the Planck's constant, 96485 C/mol.

The heat absorbed and released Q_{st} in the electrolysis process is related to entropy production and temperature:

$$Q_{st} = T(S_{out} - S_{in}) - TS_{gen}$$

In addition, the power consumption P_{st} and hydrogen yield $n_{H_2,out}$ of electrolysis process can be obtained

$$P_{st} = E J A_{SOEC} N$$

$$n_{H_2,out} = \frac{J A_{SOEC} N}{nF}$$

Where A_{SOEC} and N are the active area of SOEC and the total number of SOEC, respectively.

The experimental data are compared with the simulation results of the model, as shown in Table 1. The model voltage is in good agreement with the experimental data, which proves that the SOEC model is reliable.

Table 1 Ultimate analysis of the biomass feedstock

Voltage, V	Model		Experiment	
Current density, A/m ²	923 K	973 K	923 K	973 K
0	0.934	0.915	0.895	0.880
1000	1.131	1.033	1.064	0.972
2000	1.249	1.122	1.236	1.077
3000	1.337	1.193	1.385	1.178
4000	1.410	1.252	1.510	1.277
5000	1.474	1.303	1.580	1.360
6000	1.532	1.350	1.620	1.420
7000	1.586	1.392	1.655	1.466
8000	1.636	1.432	1.680	1.510

2.3 Modeling of biomass gasification

In the biomass gasification process, the feedstock is dried and pyrolyzed initially to release the volatile and

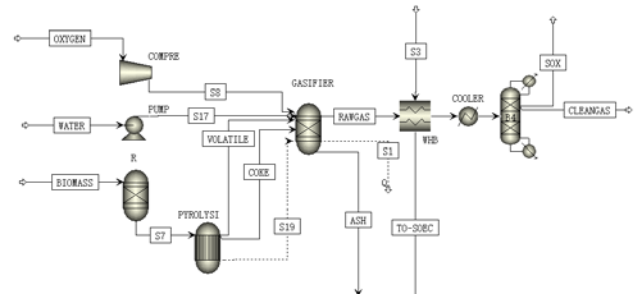
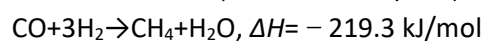
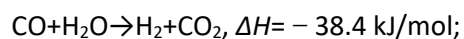
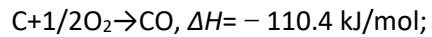


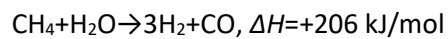
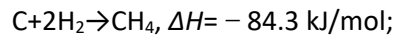
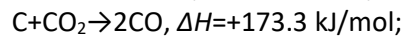
Fig. 3 Model of biomass gasification

form the coke, and then the oxidation and reduction reactions occur. The main reactions is as given below:

Oxidation reaction



Reduction reactions



Next the high-temperature raw gas from the gasifier enters the waste heat boiler (WHB) for heat recovery and desulfurization to obtain the cold clean syngas [12].

The cotton stalk from Xinjiang Province, China is chosen as the feedstock of the gasifier. Its ultimate analysis is shown in Table 2.

Table 2 Ultimate analysis of the biomass feedstock

Ultimate Analysis, wt%						
C _{ar}	H _{ar}	O _{ar}	N _{ar}	S _{ar}	A _{ar}	M _{ar}
45.01	6.094	40.04	0.601	0.043	2.686	5.525

2.4 Modeling of methanation process

The CO₂/CO co-existence methanation is commonly applied for the methane synthesis of coke oven gas. It fits

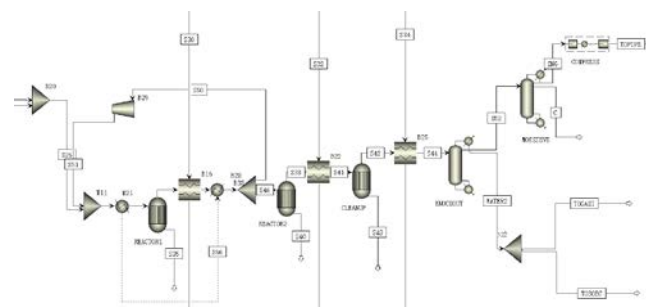
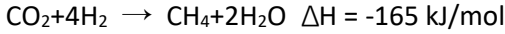
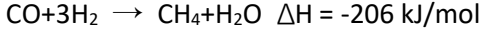


Fig. 4 Model of co-methanation

for this proposed SNG plant according to the syngas character of electrolysis and gasification process [13].

The renewable hydrogen from SOEC will mix with the clean syngas (CO+CO₂+H₂) from the gasification process and the ratio of H₂/(CO+CO₂) is set to 3. Then the methanation of CO₂/CO coexistence system will occur in the reactors in series. The CO and CO₂ will transmuted to CH₄ through following reactions.



The TREMP™ technology is adopted and the high temperature resistant nickel-based catalyst is used in the exothermic methanation process. In this process, three reactors are connected in series and the obtained natural gas will be compressed to 6.21 MPa for pipeline transportation.

2.5 Evaluation criteria and operation conditions

Some criteria are adopted to evaluate the plant performances. The methane yield Y_{CH_4} refers to the ratio of CH₄ production to the biomass feedstock.

$$Y_{\text{CH}_4} = \frac{G_{\text{CH}_4}}{G_{\text{bio}}}$$

Where G is the mass flow rate, and the subscript CH₄ and bio mean the produced methane and biomass.

η_{cold} refers to the cold gas efficiency of gasification process.

$$\eta_{\text{cold}} = \frac{G_{\text{syngas}} \text{HHV}_{\text{syngas}}}{G_{\text{bio}} \text{HHV}_{\text{bio}}}$$

The subscript syngas is the syngas product of gasification process.

η_{th} and η_{ex} represent the thermal and exergy efficiency of the SNG plant.

$$\eta_{\text{th}} = \frac{G_{\text{CH}_4} \text{HHV}_{\text{CH}_4}}{G_{\text{bio}} \text{HHV}_{\text{bio}} + \frac{P_{\text{SOEC}}}{\eta_{\text{AC-DC}}} + Q_{\text{heat}} + P_{\text{pump}} + P_{\text{compr}}}$$

$$\eta_{\text{ex}} = \frac{G_{\text{CH}_4} e_{\text{CH}_4}}{G_{\text{bio}} e_{\text{bio}} + \frac{P_{\text{SOEC}}}{\eta_{\text{AC-DC}}} + Q_{\text{heat}} \left(1 - \frac{T_0}{T}\right) + P_{\text{pump}} + P_{\text{compr}}}$$

HHV is the high heating value, P is the electric power required and the subscript SOEC, pump and compr mean the SOEC, pump and compression process. Q_{heat} is the heat requirement of the whole plant and $\eta_{\text{AC-DC}}$ is the efficiency of AC/DC conversion, equal to 0.95. e represents the exergy of different material.

The main operation parameters of the whole SNG plant are illustrated in Table 3.

Table 3 Main operation conditions

Unit	Proposed system	
SOEC	Temperature, K	973
	Cathode stream	H ₂ O 90% H ₂ 10%
	Anode stream	O ₂ 100%
	H ₂ O inlet, kmol/h	855.9
	Steam utilization, %	70
	LHV of H ₂ , kJ/kg	120210
	Exergy of H ₂ , kJ/kg	142351
Gasification	P _{gasi} , MPa	4
	T _{gasi} , K	1123
	M _{bio} , t/h	10
	Steam/Biomass _{daf}	0.5
	Oxygen/Biomass _{daf}	0.17
Methanation	Heat dissipation, %	5
	P _{in-Reactor #1} , MPa	3.24
	Ru	3
	T _{in-Reactor #1} , K	573
	T _{in-Reactor #2} , K	570
	T _{in-Clean up Reactor} , K	470
	P _{SNG} , MPa	6.21

3. RESULTS AND DISCUSSION

3.1 Parameter analysis

The changes of SOEC potential and power density with current density is shown in Fig. 5. At the same current density, with the increase of operating temperature, the cell potential and the power density of SOEC both reduces due to the decrease of polarization loss. When at 1023 K, 8200 A/m², the operation cell

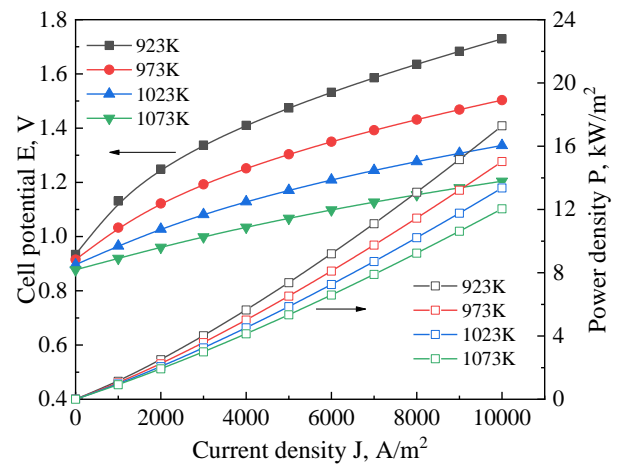


Fig. 5 Variations of cell potential and power density with current density

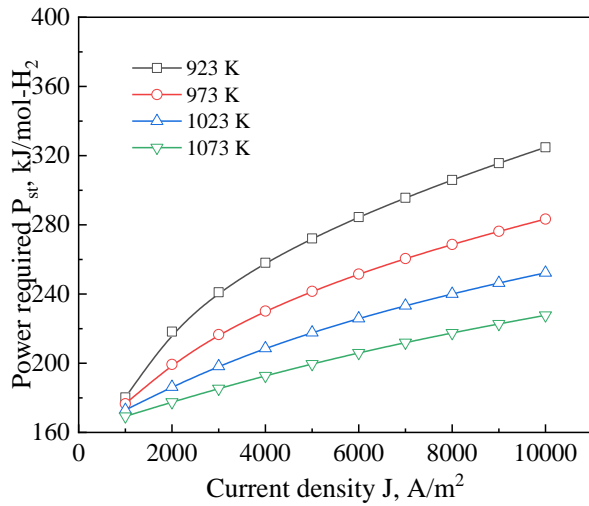


Fig. 6 Variations of power with current density

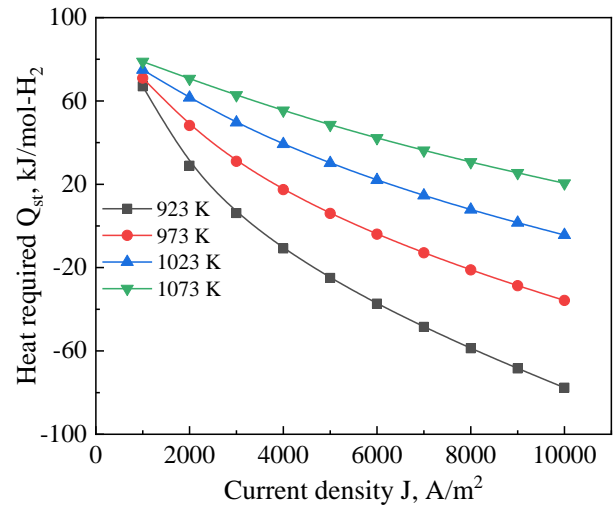


Fig. 7 Variations of heat requirement with current density

potential reaches 1.285 V (the thermal neutral voltage), which means the SOEC operates at constant temperature without generating or absorbing external heat. When the current density is less than 8200 A/m², the cell potential is lower than the thermal neutral voltage and external heat is required for SOEC to maintain the operation temperature. While operating beyond 8200 A/m², the SOEC generates heat and existing streams have higher temperature.

In Fig. 6 and Fig. 7, the actual heat consumption and power with the operating current density are evaluated. The heat consumption of SOEC increases with temperature rising while the change of power requirement is the contrary. At lower current density (< 4000 A/m²), the thermal energy demand is dominant, and after that the power requirement becomes higher, which mainly because the higher current density leads to exothermic operation, and the internal irreversibility will cause the heat generated internally.

The temperature has significant impact on the gasification energy and exergy efficiency as well. When temperature increases from 723 K to 1123 K, the gasification cold gas efficiency grows from 51% to 83%, and its exergy efficiency is from 43% to 67%. As for the co-methanation process, Ru is defined as the ratio of the mass flow rate of the circulating gas and the fresh gas entering the first methanation reactor. With the Ru increases from 0 to 9, the outlet temperature of reactor #1 decreases to 452 °C, and the power required of the recycle compressor significantly rises to 474 kW. Meanwhile with the Ru increases, more gas is recycled and the equipment capability and the cost should be larger. The recommended values of Ru is 3.0 according to the engineering experience.

3.2 Energy and exergy analyses of the SNG plant

The electricity for water electrolysis accounts for 40% energy input of the whole SNG plant approximately and biomass feedstock takes up the remaining 60%. The hydrogen from SOEC has to be compressed since the gasifier works under a high pressure. Because oxygen-steam gasification method is adopted and O₂ agent derives from the SOEC, the traditional air separation unit (ASU) is cancelled and amount of power for ASU can be saved. Meanwhile, the efficiency is higher than the air-steam gasification.

Table 4 Energy and exergy analyses of SNG plant

Energy input	Energy, kW	Exergy, kW
SOEC		
Electrolysis	43195.13	43195.13
Hydrogen compressor	3103.5	3103.50
Gasification		
Biomass	63532	71841.03
Pump	7.51	7.51
Oxygen compressor	181.6	181.6
Methanation		
Recycle gas compressor	96.24	96.24
SNG compressor	240.47	240.47
Sum.	110356.4	118665.47
Energy output		
SNG	85414.92	79586.53
Thermal/exergy efficiency,%	77.40	67.07

4. CONCLUSIONS

This work discussed a novel efficient SNG production system combined high temperature electrolysis with oxygen/steam biomass gasification. The system cancelled the conventional water gas shift process and avoided CO₂ capture consumption. The by-product and the process heat were fully recycled and utilized. The thermal and exergy efficiencies of the novel plant can reach 77.4% and 67%, respectively, which was 15 percentage points higher than the traditional biomass to methane or P2G process. The proposed SNG production system based on renewable hydrogen and biomass gasification can significantly improve the energy

efficiency of the whole plant, and the coupling process achieves zero carbon emission in the full life cycle.

ACKNOWLEDGEMENT

This paper is supported by the National Key Research and Development Program of China (Grant No. 2018YFE0208400); Project of China Electric Power Research Institute (5242002000RF): modeling and optimization of high efficiency power-to-gas system based on renewable energy.

REFERENCE

- [1] Mitigation C C. IPCC special report on renewable energy sources and climate change mitigation[J]. *Renewable Energy*, 2011, 20(11).
- [2] Kalogirou S A. *Renewable Energy*[J]. *Renewable & Sustainable Energy Reviews*, 2001, 5(4):373-401.
- [3] China Electricity Council. 2019 National Electric Industry Statistics List [R]. 2019.
- [4] GoTz M, Lefebvre J, MoRs F, et al. Renewable Power-to-Gas: A technological and economic review[J]. *Renewable Energy*, 2016, 85:1371-1390.
- [5] Schiebahn S, Grube T, Robinius M, et al. Power to gas: Technological overview, systems analysis and economic assessment for a case study in Germany[J]. *International Journal of Hydrogen Energy*, 2015, 40(12):4285-4294.
- [6] Tichler R , Bauer S . Power-to-Gas[J]. *Storing Energy*, 2016:373-389.
- [7] Clegg S, Mancarella P. Integrated modeling and assessment of the operational impact of power-to-gas (P2G) on electrical and gas transmission networks[J]. *IEEE Transactions on Sustainable Energy*, 2015, 6(4):1234-1244.
- [8] He C , Feng X , Chu K H . Process modeling and thermodynamic analysis of Lurgi fixed-bed coal gasifier in an SNG plant[J]. *Applied Energy*, 2013, 111(11):742-757.
- [9] Chandel M, Williams E. Synthetic Natural Gas (SNG): Technology, environmental implications, and economics. Climate Change Policy Partnership, Duke University, Durham, NC; 2009. available at: <https://nicholasinstitute.duke.edu/sites/default/files/publications/natgas-paper.pdf>.
- [10] Alzahrani A A, Dincer I. Thermodynamic and electrochemical analyses of a solid oxide electrolyzer for hydrogen production[J]. *International Journal of Hydrogen Energy*, 2017:21404-21413.
- [11] Diethelm S, Herle J V, Montinaro D, et al. Electrolysis and co-electrolysis performance of SOE short stacks[J]. *Fuel Cells*, 2013, 13(4):631-637.
- [12] Wang D , Li S , He S , et al. Coal to substitute natural gas based on combined coal-steam gasification and one-step methanation[J]. *Applied Energy*, 2019, 240:851-859.
- [13] Yongtian Y, Yanhua Y, Xu Z, et al. Methanation of three different reaction systems of carbon oxides [J]. *Chemical Industry and Engineering Progress*, 2014.