Economic Analysis of Syngas Production by Coupling Coal Gasification Wastewater Treatment and Solid Oxide Cell System

Peng Wang, Qiao Zhang*

School of Chemical Engineering and Technology, Xi'an Jiaotong University, Xi'an, Shaanxi 710049, China
qzhang1986@xjtu.edu.cn

Coal is the main energy source in China, and coal chemistry wastewater treatment has always been a research hotspot. For the large amount of concentrated organic wastewater, high cost caused by complex treatment process is the key issue. In this paper, the fixed bed Lurgi gasifier wastewater coupled with solid oxide cells (SOCs) system is employed for its treatment. Based on Aspen Plus software, the purified methane from marsh gas is sent to solid oxide fuel cell (SOFC) for electricity generation. While carbon dioxide separated from biogas purification and acid gas removal enters solid oxide electrolysis cell (SOEC) to produce syngas by co-electrolysis with steam.The simulation results show that the efficiency of SOFC is 48.5 %. The generated electric energy can be used for auxiliary equipment in wastewater treatment and SOEC to produce syngas, recycling of carbon dioxide. SOEC with wind power of 12.84 MW is enough to treat the waste gas generated by the 1,100 t/h Lurgi gasification wastewater treatment process. Economically, the cost of producing syngas, with high purity and fewer contaminant contents, is about 0.126 $/Nm^3. It can be used to produce various chemicals, so as to further improve the economic benefit of wastewater treatment.

1. Introduction

Coal chemical industry is a manufacturing process with high pollution, high energy consumption and massive water consumption and wastewater discharge. As stricter environmental protection requirements, wastewater treatment should be emphasized in coal chemical industry, and the cost reduction of wastewater treatment becomes the key problem.

Using the fixed-bed Lurgi gasifier as an example, gasification of 1 t of coal will produce about 0.8 - 1.1 m^3 of wastewater (Wang, 2014). Pollutants in wastewater are high ammonia nitrogen, salt and concentrated organic species, containing refractory and toxic substances, such as phenol, aromatic hydrocarbon, heterocyclic compound and cyanide, etc. Generally, it needs to go through particles and oil removal, phenol ammonia recovery, biochemical treatment and advanced treatment to satisfy the reuse standard. At present, the main method to reduce cost is to develop new processes or optimize the original process. The representative processes of phenol ammonia recovery include deacidification-deamination-e xtraction dephenolization, single tower pressurized side line extraction-extraction dephenolization process of South China University of Science and Technology and phenomenolvan process of Sasol company in South Africa. As for the previous one, acid gas (CO$_2$, H$_2$S) in wastewater can be removed and ammonia can be recovered. The chemical oxygen demand (COD) is generally in the range of 2,000 – 3,000 mg/L(Cui, 2017). For biological treatment units, it usually includes activated sludge process, A^2/O and enhanced anaerobic process, etc. Li (2017) uses the effluent reflux anaerobic fermentation (AF) process to treat coal-to-natural gas wastewater and obtains methane by comparing the treatment effects of up-flow Anaerobic Sludge Blanket (UASB) reactors under different operating processes. In industry, the acid gas and biogas produced by wastewater are treated by sulfur recovery and co-generation. Such technology will generate a large amount of CO$_2$, and the efficiency of cogeneration is generally low at 35 %, causing a certain waste of resources and energy.

Conventional solid oxide fuel cells (SOFC) are divided into solid oxide full cell and solid oxide electrolysis cell. The generation efficiency of SOFC without restriction of carnot cycle is higher than that of co-generation and

able to reach the requirement of commercial application. Minutillo et al. (2014) simulated SOFC and molten carbonate fuel cell (MCFC) using Aspen Plus to predict the state performance of cells under different conditions. Mehrpooya et al. (2019) established and analyzed the combined cooling heating and power system based on SOFC. However, SOEC is still in the pilot stage, and there is certain distance from application. Ni (2012) established a mathematical model of co-electrolysis of CO_2 and H_2O based on SOEC. Samavati (2018) designed and analyzed the process of SOEC and biomass gasification to produce Fischer-Tropsch diesel. A new way for energy-efficient utilization and CO_2 reuse has been provided by the development of SOCs system. In order to solve the high cost problem of coal gasification wastewater treatment, the Lurgi Gasifier gasification wastewater and existing treatment process are coupled with the solid oxide cells (SOCs) system to produce syngas. The characteristic of the process is to select the appropriate technology for biogas decarbonization and couple the SOCs system with the coal gasification wastewater treatment process for reasonable utilization of CH_4 and CO_2. All processes are established in Aspen Plus (Aspen Plus, 2017), and its economic analysis was carried out on the production of syngas under different current densities by SOEC in thermal neutral mode. The cost of coal gasification wastewater treatment can be reduced by realizing the resource utilization of the wastewater treatment.

2. Process description

The block flow diagram of coupling solid oxide cell system and coal gasification wastewater treatment is shown in Figure 1. Phenol ammonia recovery, enhanced anaerobic process, desulfurization unit, biogas purification unit, SOFC and SOEC are included in this coupling system. After dedusting and deoiling, the wastewater first enters the phenol ammonia recovery unit, which can remove acid gas from the wastewater, recover valuable ammonia, and obtain crude phenols by the extraction process. Wastewater after the regeneration treatment of the extractant enters the enhanced anaerobic device of the biological treatment and generates biogas. By subsequent treatment, the water quality can reach the reused water standard. Methane with high purity is obtained by the purification unit and sent to SOFC for high-efficiency power generation. While sour gas from phenol ammonia recovery is mixed with CO_2 separated from biogas purification after sulfur recovery unit, and sent to SOEC as cathode feed together with steam to produce syngas through co-electrolysis reaction under external wind energy. The electric energy produced by SOFC can be used by auxiliary equipment and SOEC.
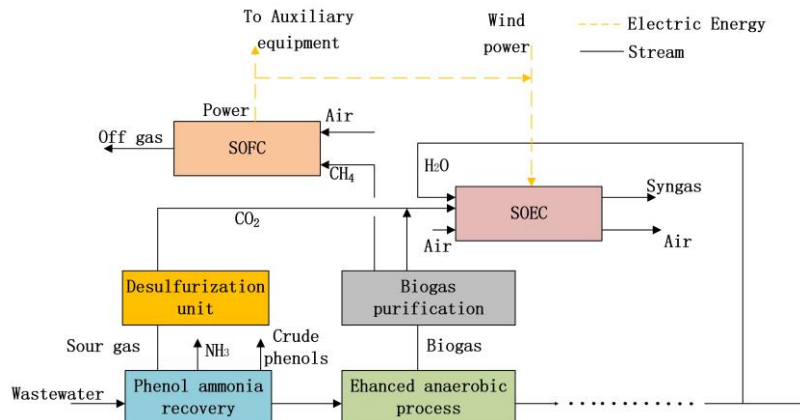

Figure 1: Block flow diagram of coupling solid oxide cells system and coal gasification wastewater treatment

3. Process modeling and results

The established process is modeled and simulated in Aspen Plus (Aspen Plus, 2017). In the wastewater treatment process, the interaction parameters from Aspen Plus acid water package (esouro) are calculated, and ELECNRTL is selected as the physical method to obtain accurate simulation results (Aspen Plus, 2017); biogas purification unit is also used with the same interaction parameters and physical method, and Peng-Robinson is employed for the solid oxide cells (SOCs) system. The wastewater is produced by gas washing process of coal gasification. Due to the presence of toxic inhibitors, large flow fluctuation and the software does not have the ability to simulate biological reactions, the biogas production and components are estimated based on the anaerobic experiment conducted by Li (2017). The phenol ammonia recovery process adopts the single tower pressurized side line ammonia extraction + MIBK process of South China University of technology, and the

simulation of single tower pressurized side line ammonia extraction process is shown in Figure 2 (Cui, 2017). Process flow chart is as follows, a total of 11 sets of equipment are required to treat 1,100 t/h wastewater.
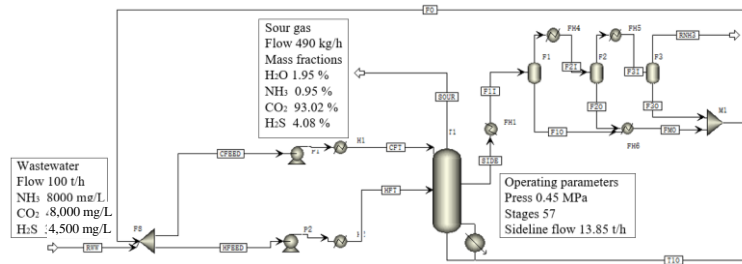

Figure 2: Process flow chart of single tower pressurized side line ammonia extraction with Aspen Plus

Due to large-scale decarburization and desulfuration devices often existing in coal chemical plants, the simulation of acid gas in wastewater treatment is ignored and CO_2 recovery is assumed to be 1. High-purity methane and separated carbon dioxide are obtained by high pressure water scrubbing absorption process (Cozma, 2015). Absorption and desorption towers use Radfrac modules. The calculation of the amount of fresh water is done through Design-spec to make concentration of enriched methane up to 95 %. Unlike original simulation process, the pressure of flash tank (Block FCO2) is set to 0.1 MPa to obtain CO_2 instead of releasing it to the atmosphere through desorption tower.
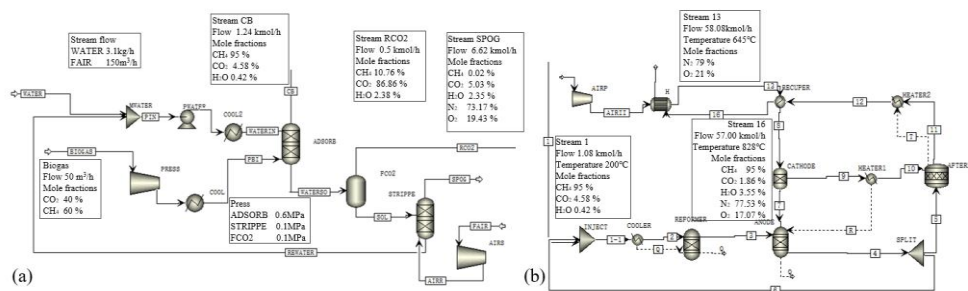

Figure 3: Process flow chart of (a) high pressure water scrubbing absorption process and (b) SOFC

Figure 3 shows the simulation of high pressure scrubbing absorption process and SOFC, and the relevant stream data is shown in this figure. For the simulation of solid oxide fuel cell, according to Zhang et al. (2005) and Minutillo et al. (2014) , the feed flow rate of SOFC is calculated by Aspen Plus™. Design-spec function to make the output power reach 120 kW. Voltage, current and other parameters are calculated using FORTRAN code in Aspen Plus. While remaining methane after biogas purification is sent to the cathode of SOEC. The simulation results are shown in Table 1. Its stack exhaust gas can exchange heat with the anode and cathode feed streams without external energy, and fuel cell efficiency is at 48.5 %.

Table 1: SOFC model simulation results

	Model simulation results
Voltage (V)	0.64
Current density (A/cm^2)	0.19
Output Power (kW)	120
Pre-reform outlet temperature (℃)	535
Efficiency(LHV)	48.5 %

The simulation of SOEC also needs to use Aspen Plus™ Design-spec function and FORTRAN code to determine the relevant data (Samavati, 2018). SOEC simulation flow chart is shown in Figure 4.
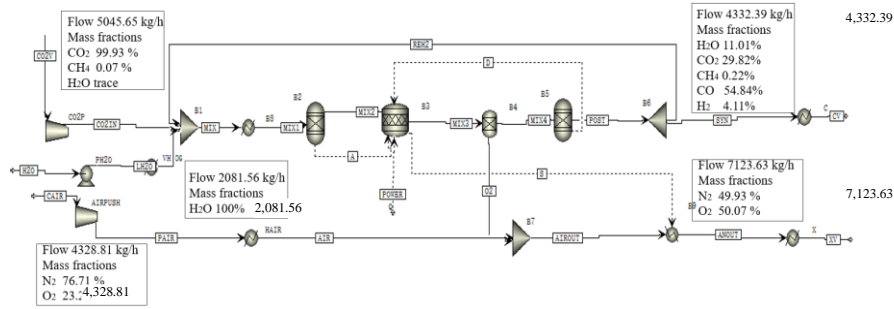
Figure 4: Simulation flow chart of SOEC (thermal neutral) at current density = 0.7 A/cm^2 with Aspen Plus

Table 2: Related results of SOEC (thermal neutral) at current density = 0.7 A/cm^2

Current density (A/cm^2)	Temperature (℃)	Area (m^2)	Syngas (Nm^3)	Effective fraction (mole %)	Voltage (V)	ASR (Ω × cm^2)	Heat Exchanger Area(m^2)
0.7	766.78	1,337.64	5,145.08	75.42	1.38	0.58	3,086.51

Table 2 shows the results when current density is 0.7 A/cm^2 and the outlet gas temperature of SOEC (thermal neutral mode) is cooled to 200 °C for heat integration. Considering the location factors, the auxiliary equipment (including pumps and compressors) of biogas purification, SOFC, and SOEC units are provided by part of the power output by SOFC, and the rest is sent to SOEC. It can be seen that the effective mole fraction of syngas increases with the increase of current density in the thermal neutral mode, and the effective mole fraction of syngas on dry-basis is more than 85 %. Table 3 summarizes the energy consumption of each unit.

Table 3: Energy consumption of each unit

Energy consumption (kW)	Deacidification and deamination unit	Biogas purification	SOFC	SOEC
Cold utility	8,770	6.28	—	—
Heat utility	21,650	—	—	1,590
Electric energy	269.19	9.92	4.17	12,878.91

4. Techno-economic analysis

In order to directly evaluate the economy of syngas production, only the biogas purification and solid oxide cells system units are calculated, but not the wastewater treatment process and desulfurization unit cost. Referring to the general process design principles (Seider et al., 2004) and the research on solid oxide electrolysis-based systems (Samavati et al., 2017), this economic evaluation includes the total capital investment and depreciation involving fixed capital, as well as utility costs, operation & maintenance costs and feed costs belonging to current capital.

4.1 Total capital investment

In this study, the total capital investment includes bare module investment of some equipment, SOCs investment, site preparation cost, contingencies and contractor' s fee, land cost and plant startup cost. The cost of auxiliary equipment in the biogas purification unit and solid oxide cell system is estimated by Aspen process economic analyzer V8. Stainless steel floating head heat exchangers are selected in the SOCs system, and the cost is calculated according to Seider et al. (2004).

The cost of SOFC stack is calculated as 140 $/kW (Yao, 2019), and the price of SOEC stack is estimated by Eq(1) (Samavati et al., 2017).

$$C_{SOEC} = C_{base} \left(\frac{ECC}{BCC} \right)^X \tag{1}$$

C_{base} represents the basic cost of SOEC, taking 0.00055 $/m^2; ECC represents the activation area of SOEC simulated in the previous section and BCC is the basic area of SOEC based on scaling parameters, BCC is 1 m^2; x is the cost proportion factor, taking 1 for SOEC.

To explore the impact of current density on investment, Figure 5 shows the total capital investment of the system at different current densities. The direct investment in equipment accounts for 76.74 % of the total capital investment; the contingencies and contractor' s fee is the second, accounting for 16.52 %; the proportion of site preparation cost, land cost and driving cost is similar. All in all, the investment of each part will increase with the increase of current density. Comparing the investment when the current density is 0.7 A/cm^2 and 0.9 A/cm^2, the increase of capital cost mainly comes from heat exchangers, whose investment accounts for 264 % of the total increase.
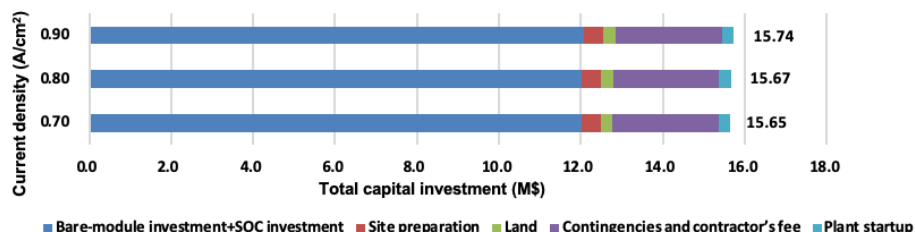

Figure 5: Total capital investment of coupling coal gasification wastewater treatment and SOCs system

4.2 Operating cost

Utility costs include two parts: fired heat and wind power. For fired heat, unit cost can be extracted from Aspen simulation results, while wind power is purchased directly from the power company. The 2020 guidance price for wind power issued by the National Development and Reform Commission is 0.04 - 0.07 $/kWh in 2020. According to the location of the coal chemical plant, wind power is calculated as 0.04 $/kWh. Steam used in the design process is supplied by the wastewater after treatment. In order to facilitate the subsequent calculation, the fresh water price of 0.428 $/t is used for economic calculations. Operation and maintenance costs are estimated at 4 % of total capital investment.

4.3 Net present value

NPV is a more comprehensive and scientific economic evaluation method, which considers the time value of capital and the economic benefits of the project after the investment is recovered in the whole life cycle. In order to calculate NPV, the cash flow in the whole life cycle shall be calculated according to Eq(2).

$$(2)$$

In above formula, tax is tax rate, s is annual sale revenue, C is annual production costs, and D is depreciation. The annual sales revenue is calculated according to the price of coal to carbon monoxide of 0.163 $/m^{3}CO (Jing, 2006), and hydrogen is roughly estimated at 2,857 $/t. Depreciation life is 15 y, tax rate and residual value rate are 15 % and 5 % (Yao, 2019).

Under the above three current densities, the net present value in the eighth year changes from negative to positive, with little overall difference; when current density is 0.9 A/cm , the recovery cost is the slowest. It is worth mentioning that when operating cost is taken as evaluation standard, the depreciation cost is no longer included. The cost per unit volume of syngas is about 0.126 $/Nm , which increases slightly as increase of current density.
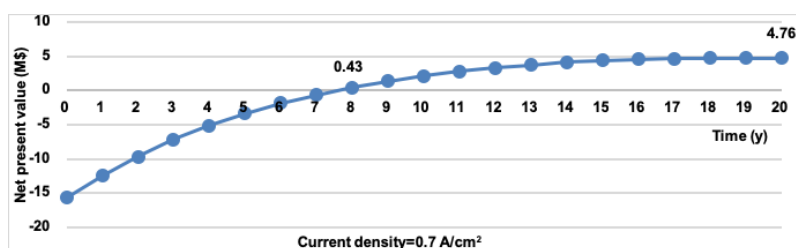

Figure 6: Net present value of syngas produced by coupling wastewater treatment and SOCs system

Figure 6 shows the NPV curve when the current density is 0.7 A/cm . According to the analysis of the wastewater treatment system, cost of treatment of wastewater by the phenol ammonia recovery system is 0.028 $/t (Cui,

2017). After coupling the SOCs system, the cost of the phenol ammonia recovery system can be reduced to 0.001 $/t in 20 y. In general, when SOEC is designed with a small current density in the thermal neutral mode, although its active area is increased and the cost is higher, the one-time investment of the system is the least and the recovery of funds is fastest. The reason may be that the investment cost of biogas purification and SOFC is fixed, the reaction equilibrium temperature corresponding to the low current density is also low under the thermal neutral mode; the cost of SOEC equipment increases, but the investment cost of heat exchanger which affects more is reduced.

5. Conclusions

Based on the combination of traditional biogas and SOFC, a new coupling system of coal gasification wastewater treatment and SOCs is proposed, and the effect of different current densities on the cost of syngas production in the thermal neutral mode of SOEC is discussed. At the same time of improving efficiency of biogas power generation, we can make full use of the value of CO_2 as a resource to produce syngas with high purity and fewer contaminations, which is further processed into high value-added products. SOEC with wind power of 12.84 MW is enough to treat the waste gas generated by 1,100 t/h Lurgi gasification wastewater treatment process. Technical and economic analysis shows that as the increase of current density, the total capital investment increases; heat exchangers have greater impact on total investment than SOEC. This study can provide a reference for the design of SOCs system integrated with waste gas and wastewater treatment unit. The impact of high-temperature waste gas utilization and subsequent fine chemicals production on the system is also worth further discussion.

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