

# Developing a novel gasification-based sludge-to-methanol utilization process and exergy-economic-environmental (3E) analysis

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## Abstract

Sewage sludge is a common waste from wastewater treatment plants in Hong Kong and conventional treatment ways like the landfilling method has been gradually abandoned considering the environmental impacts and costly land. Searching for integrating processes to well treat the sludge simultaneously achieve the energy recovery can be a good alternative way. Therefore, process design and simulation of a gasification-based sludge to methanol (STM) production was proposed in this work. It considered major subsections such as gasification, power generation, absorption, methanol synthesis, and distillation. Followed by the sensitivity analysis to optimize operational parameters, economic, environmental, and exergy (3E) analysis of STM process was conducted. The economic estimation revealed the methanol production cost is 579.62\$/ton and the sludge disposal cost is 39.58\$/ton. The calculated internal rate of return (IRR) of the process is 10.20% and 5.48%, respectively with a subsidy of 20\$/ton sludge and without any support. Moreover, it was found the greenhouse gas (GHG) emission was 3.21 kg eq.CO<sub>2</sub>/kg methanol and overall exergy efficiency was 57.31%. The current work implies the great potential of STM treatment strategy, which can serve as a reference for the future sludge treatment planning.

**Keywords:** Sludge gasification, Waste to energy, Exergy analysis, GHG emission, Internal rate of return

## 1. Introduction

Huge amount of wastewater in modern city has raised a growing attention on the study of its sustainable treatment [1]. Sewage sludge is a residue solid-liquid mixture that generated from such wastewater treatment processes. It contains a high proportion of moisture and thereby the volume is large. The direct objectives of sludge treatment are to reduce the volume and to stabilize the material by destructing some pathogenic microorganisms [2]. In the case of Hong Kong, it receives about 380 kilo tons of sewage sludge per year from 11 collection centers [3]. For the goal of sustainable development of Hong Kong, exploration and valorization of sewage sludge is necessary. Liu et al [4] has reviewed the developments of utilizing sludge to produce hydrogen and compared the specified technologies in terms of their advantages and drawbacks. From the review, biological conversion has the cost superiority however it cannot substantially reduce the sludge volume. On the other hand, gasification method has the superiority in technical readiness level and pollution alleviation among thermochemical processes, it has been widely studied before with the expectation of achieving the circular economy [5]. Gasification technology can convert waste resources into a syngas mixture containing carbon monoxide (CO), hydrogen (H<sub>2</sub>), carbon dioxide (CO<sub>2</sub>), vapor water, methane (CH<sub>4</sub>) and other light combustible hydrocarbons (C<sub>x</sub>H<sub>y</sub>) which could serve as precursors to produce waste-based fuels or value-added chemicals [6, 7]. Owing to the complex mechanism of the gasification, some experimental investigations have been carried out to contribute more on the kinetics of syngas production [8, 9] or the feedstock decomposition [10, 11]. Followed by the laboratory results, conceptual design by process simulation can be applied as a representative scale up way to explore the real conditions during the establishment of gasification plants [12]. For example, Hantoko et al [13] investigated the supercritical water gasification (SCWG) of sewage sludge for hydrogen production by using a thermodynamic equilibrium model, in which various operation factors were analyzed in detail. Zhang et al [14] reported an enhanced SCWG integrated system for sludge treatment and the thermodynamic analysis was conducted accordingly using sensitivity analysis. Different from the gasification simulation using Aspen Plus platform, another suitable mathematical model considering the thermodynamic equilibrium was formed in Matlab platform by Bijesh et al [15]. They explored the effects of temperature and equivalence ratio on the molar distribution of gas products. Notably, Yang et al [16] established a commercial biomass intermediate pyrolysis poly-generation process using

1 Aspen simulation tool and shows the prospective contribution of biomass utilization to China's carbon  
2 reduction goal. Although the literatures made some contribution on the gasification model and  
3 operational conditions, limited work on the utilization of the sewage sludge in Hong Kong has been  
4 analyzed so far. Whether the gasification treatment of sewage sludge can be economically feasible for  
5 future policy making in Hong Kong is worth to be studied.

6 One of the downstream products of gasification syngas was methanol, which has been seen as a  
7 promising fuel and essential chemical raw material which can be used to produce olefins, acetic acid,  
8 formaldehyde, and methyl tertiary butyl ether (MTBE) in the chemical industry [17]. Generally, two  
9 different pathways for the methanol production are biological and thermochemical process, in which  
10 thermochemical conversion using primary energy resources like natural gas (NG) as the raw material  
11 to achieve the methane reforming was mainly applied currently [18]. However, considering the huge  
12 carbon emission in conventional production and nonrenewable nature of coal and NG, developing  
13 alternative processes to achieve the methanol production from other feedstocks is a promising  
14 approach. For example, Hernández et al [19] make an optimization of the biogas-to-methanol synthesis  
15 process based on mathematical models. Optimization results with the environmental and economic  
16 objectives shown the biogas composition should be around 50% CH<sub>4</sub> and 47% CO<sub>2</sub>. Similarly, Santos  
17 et al [20] established mathematical model of the biogas-to methanol synthesis process and comparably  
18 studied various biogas sources namely landfill, palm oil effluent, corn cobs and sorghum formation  
19 with the maximum methanol amount. The above investigations did not study the biomass-to-biogas  
20 conversion and the major focus was the optimization of biogas-to-methanol synthesis. In regard of this,  
21 Puig-Gamero et al [21] proposed the thermochemical conversion process of methanol production by  
22 integrating pine gasification, syngas cleaning and methanol synthesis sections. It conducted the  
23 analysis of the effects of different variables on the final syngas composition.

24 As the gasification modelling gets deeper, exergy, economic and environmental (3E) analysis of  
25 the systematic process of methanol production from waste or biomass has been gradually investigated,  
26 which aimed at validating the technology prospect of waste valorization. For instance, Im-orb et al [22]  
27 carried out a thermodynamic analysis of bio-methanol production from oil palm residues using a  
28 process model developed in Aspen Plus and it was found the maximum energy and exergy efficiencies  
29 were 38.57 and 25.44%, respectively. Moreover, the thermodynamic analysis were extended in the  
30 waste valorization processes to produce synthetic fuel [23], power [24] and ethylene glycol [25]. On

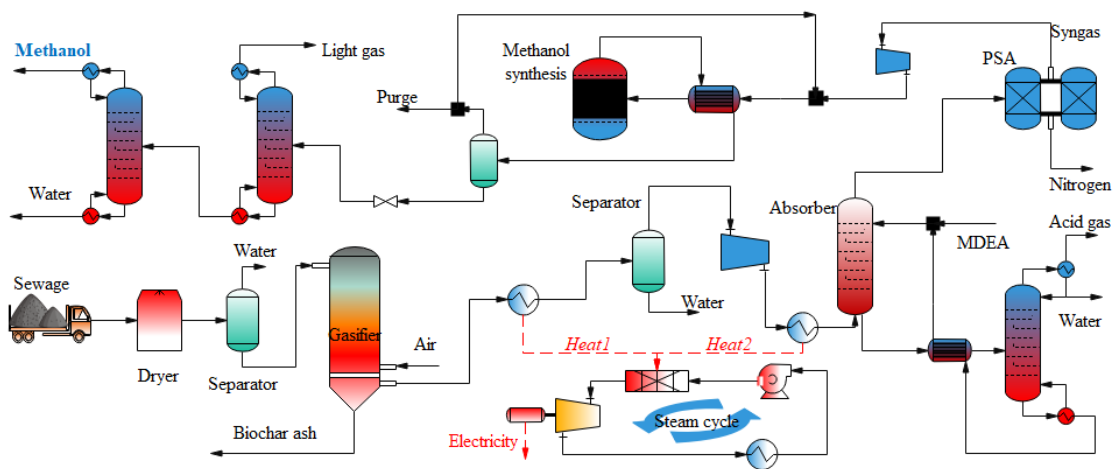
1 the other hand, economic and environmental analysis of such waste valorization has attracted more  
2 attention. For example, Menin et al [26] have modelled two biomass gasification-based processes  
3 namely the standalone bio-methanation of syngas (SAB) and the integrated biomethane and bio-  
4 methanol (IBB) production. Qi et al [27] made a conceptual design of novel municipal sludge-to-  
5 hydrogen fuel production based on plasma gasification, in which they analyzed the technical, economic  
6 and environmental performance and the economic results has shown a huge potential of sludge-to-  
7 hydrogen in sludge treatment. Alves et al [28] finished the development of a techno-economic analysis  
8 for a small-scale gasification plant for the production of hydrogen or electricity power. In terms of  
9 obtaining the bio-char products, Medina-Martos et al [29] comparably studied the sludge hydrothermal  
10 carbonization process however the economic cost was found to be 42% higher than that of standard  
11 lone anaerobic digestion. The techno-socio-economic analysis of the sludge-to-biooil pyrolysis  
12 conversion was conducted systematically by Shahbeig et al [30]. More recently, a comprehensive  
13 analysis was finished for the bio-methanol production from palm wastes steam gasification which  
14 applied the carbon capture technology [31]. the calculated profit was around 193.5\$/tonne based on  
15 the assumption of methanol selling price 430\$/tonne and carbon reduction was around 57.25 tonnes  
16 CO<sub>2</sub> -e/h. From the literature review, although economic and environmental evaluation on the sludge-  
17 to-hydrogen, sludge-to-biooil, sludge-to-biochar, and biomass-to-methanol has been studied, few  
18 research have focused on a systematical simulation and the comprehensive analysis from economic,  
19 environmental and exergy aspects of sludge-to-methanol (STM) process.

20 Therefore, according to the requirement of sludge treatment in Hong Kong, this work intend to  
21 investigate the gasification-based valorization method to obtain methanol production from waste  
22 sewage sludge. The main contributions of this study are: 1) a systematic process from raw sludge to  
23 methanol conversion was proposed which involves the gasification treatment and downstream  
24 methanol production, 2) the key operational parameters of the STM process were optimized  
25 sequentially based on the rigorous simulation by Aspen Plus, and 3) comprehensive analysis were  
26 conducted from 3E aspects to provide some suggestions and implications. Specifically, the integrated  
27 STM process was mainly comprised of gasification, power generation, absorption, methanol synthesis,  
28 and distillation. The gasification model was established through the restrict thermodynamic  
29 equilibrium of typical gasification reactions and the adjustment of temperature approach. Then,  
30 gasification gases distribution in simulation was compared with that in literatures to make a validation

1 of the gasification model. Followed by that, the sensitivity analysis was finished to investigate the  
 2 influence of key factors on the syngas yield and composition. According to the systematic built-up  
 3 process after optimization, the economic estimation has shown the feasibility of sludge gasification in  
 4 Hong Kong despite of some subsidy. We also carried out the life cycle GHG emissions to evaluate the  
 5 environmental impacts. A detailed exergy flow calculation was carried out to identify the energy  
 6 bottleneck and provide guidelines for further process improvement.

## 7 2. Methodology

### 8 2.1 Process simulation



9  
 10 Fig.1 Systematic gasification utilization process of sewage sludge for methanol production

11 The simplified process flowsheet was shown in **Fig. 1**. It is assumed that the proposed process is  
 12 a conceptual design and is the major sections of a real-world waste-to-energy plant. Additional pips  
 13 and valves were not displayed in the process flowsheet. Five major operation units were involved such  
 14 as the air gasification, steam-cycle power generation, gas removal, methanol synthesis and methanol  
 15 distillation separation. The sewage sludge was generated during treatment of wastewater from different  
 16 sites in Hong Kong. After collecting the sewage sludge in a centralized disposal site, the sludge was  
 17 dried under the atmosphere of high-temperature steam. Most of the bacteria and virus would be killed  
 18 and then moisture be removed to adjust the water flowrate into the gasifier. Gas mixture emitted from  
 19 the downdraft gasifier was a heat source that can evaporate the high-pressure water into steam and  
 20 achieve power generation. This integrated process can reduce the electricity consumption in other  
 21 operations. Flue gas contained  $H_2S$  gas which is highly toxic and can deactivate catalysts in the  
 22 methanol synthesis. Therefore, acid gas must be absorbed and stored to avoid the damage. Then inert

1 nitrogen gas can be separated through pressure swing adsorption (PSA) technology. Nitrogen gas is  
 2 popular and valuable product in the gas market. Through the gas cleaning section, purified syngas with  
 3 main components CO, H<sub>2</sub>, CH<sub>4</sub>, CO<sub>2</sub> and H<sub>2</sub>O were sent into the synthesis reactor to synthesize  
 4 methanol. Eventually, distillation process achieved the separation of methanol product with 99.9 mol%.  
 5 Complete process simulation was built up in the Aspen platform with their detailed model  
 6 specifications, which has been shown in **Fig. A1** and **Table A1**, respectively. Detailed demonstration  
 7 of each simulation is given in the following subsections.

### 8 2.1.1 Gasification process

9 Similar to the biomass gasification, the complete simulation of waste sludge gasification can be  
 10 seen as three stages namely drying, pyrolysis and gasification stages. The dryer was modelled using a  
 11 Rstoich reactor block which can convert a part of nonconventional sludge into water according to the  
 12 conversion reaction (see **Eq. (1)**).



14 Since the feedstock of sewage sludge was introduced in Aspen Plus as a nonconventional  
 15 component, before going into the gasification reactor, it should be decomposed into its elements  
 16 namely the conventional components. Therefore, another RYield reactor unit was applied to simulate  
 17 the pyrolysis process according to the ultimate analysis of the sewage sludge. This is NOT a true  
 18 standalone reactor, but an integral part of the gasification reactor which provides the simplicity to  
 19 determine various species like solid carbon, hydrogen, nitrogen, oxygen, sulphur, bounding water and  
 20 ash. Then, inert ash was separated by a cyclone separator before they were introduced into the  
 21 gasification reactor. Based on the experimental research, **Table 1** has shown the industrial analysis  
 22 composition of sewage sludge in detail.

23 Table 1. The typical composition of sewage sludge in Hong Kong [32]

Proximate analysis (wt%)		Ultimate analysis (wt%)	
Moisture	64.1	C	47.54
Volatile matter	75.3	H	7.99
Fixed carbon	1.3	O	18.55
Ash	23.4	N	2.02
		S	0.5

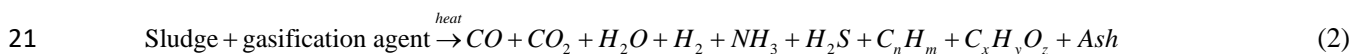
24 A common method was used in this work for gasification modelling by estimating thermodynamic  
 25 equilibrium through calculation of Gibbs energy minimization. However, real operation cannot reach

1 the idea equilibrium condition. Therefore, the temperature approach was adjusted to obtain model  
 2 predictability by restricting some predefined reactions in the Gibbs reactor [33, 34]. In this work,  
 3 specified outlet components and restricted reactions have been given in **Table 2**. Notably, the number  
 4 of involved atoms plus that of linearly independent reactions must equal to the total number of products.

5 Table 2. Specified components and restricted reactions of the gasification Gibbs reactor [33]

Involved atoms	Restricted reactions	Specified output products
C	R1: $C + CO_2 \rightarrow 2CO$	H <sub>2</sub> C CO CO <sub>2</sub> H <sub>2</sub> O NH <sub>3</sub> N <sub>2</sub> H <sub>2</sub> S CH <sub>4</sub> O <sub>2</sub>
H	R2: $C + H_2O \rightarrow CO + H_2$	
O	R3: $CO + H_2O \rightarrow CO_2 + H_2$	
N	R4: $CH_4 + H_2O \rightarrow CO + 3H_2$	
S	R5: $C + O_2 \rightarrow CO_2$	

6 Additionally, some basic assumptions were made to simplify the downdraft gasifier before  
 7 conducting the simulation [35]: 1) the gasification system is at steady state isothermal condition, 2)  
 8 the nitrogen and sulphur were fully converted into NH<sub>3</sub> and H<sub>2</sub>S, 3) ash is inert and tar formation are  
 9 neglected because of the low content in the outlet gas stream from downdraft gasifier, 4) char only  
 10 contains carbon and ash. PR-BM physical model was used in the gasification stage since it can well  
 11 estimate the properties of such hydrocarbon and light gases in a fairly high temperature condition.  
 12 Components like ash and the sewage sludge have been set as non-conventional component. Enthalpy  
 13 and density estimation of non-conventional sludge and ash was finished using HCOALGEN and  
 14 DCOALGT model. The HCOALGEN model includes several correlations for the heat of combustion,  
 15 formation, and heat capacity which can be found in the Aspen Help files. In the present work, the  
 16 setting of HCOALGEN model was 6-1-1-1 which means the heat of combustion is user defined and  
 17 the heat of combustion of sewage sludge selected in this work is 25.63 MJ/kg based on the  
 18 experimental result [32]. It is worth noting that gasification is not autothermal and additional fuel must  
 19 be supplied to meet the gasification requirements. Through the three steps, a sewage sludge gasification  
 20 process can be understood by a universal equation (see **Eq. (2)**).



1 Aromatic tar ( $C_xH_yO_z$ ) components generated during the intermediate low-temperature pyrolysis  
2 was ignored since downdraft gasification is recognized to produce insignificant such contents [36].  
3 Independent gasification reactions that considered were listed in **Table A2** in the **Appendix. A** [37].  
4 The downdraft gasifier was operated in the atmospheric pressure and the temperature of dryer,  
5 pyrolysis and gasification were 101, 500 and 800 °C [38].

### 6 2.1.2 Power generation

7 As shown in **Fig. 1**, the high-temperature gas from gasifier can be used to vaporize water and  
8 achieve the power generation. This integrated gasification that combined cycle can be found more  
9 detail in the studies [39, 40]. Herein, steam cycle was applied owing to the availability of water. It  
10 consists of a heat recovery evaporator, steam turbine, pump, and condenser. There are two steam cycle  
11 processes for the power generation. The first heat comes from the raw syngas produced from downdraft  
12 gasifier. And the second heat exchanger is used to recover the inner energy of compressed syngas  
13 followed by connecting an absorption column. To make the power generation process converge,  
14 following procedures were conducted. Firstly, a suitable feed flowrate should be found with  
15 appropriate heat-exchange conditions where no cross-temperature exists. In detail, a design  
16 specification was set to vary the feed flow rate to attain a suitable cold stream outlet temperature in  
17 which the outlet steam has been completely vaporized. Then, pressures of discharged stream from  
18 pump and turbine were determined through the sensitivity analysis with the aim of maximum power  
19 generation. Considering the complete condensation of recycled steam from turbine and the required  
20 minimum temperature difference of 5°C between it and utility water, the discharged pressure of turbine  
21 cannot be too small.

### 22 2.1.3 Acid gas removal process

23 The composition of inlet syngas to the absorber column has been given in **Table 3**. It contains  
24 undesirable compounds like  $H_2S$  and  $NH_3$  which have harmful effects on the downstream methanol.  
25 Therefore, this section aims to separate those acid gas and the remaining gas can be introduced into  
26 the methanol synthesis reactor. The process described in this work has shown a typical absorption  
27 system. More detailed information about the simulation of the amine solvent absorption can be found  
28 in the tutorial book [41]. In this work, absorption model was built up on the chemical reactions basis  
29 with a methyl diethanolamine (MDEA) solution. MDEA as a tertiary ethanolamine was selected due  
30 to its higher selectivity absorption of  $H_2S$  compared to other amine solvents [42]. Another advantage



1 of using MDEA is the lower heat of regeneration process than other amines, which is helpful in  
 2 reducing the total energy consumption of the desorption column.

3 Table 3 The composition of raw syngas that flow into the absorption column

Composition	Mole flowrate (Kmol/h)	Mole fractions
H <sub>2</sub>	1109.8203	0.5264
CH <sub>4</sub>	2.6474	0.0013
CO	441.0581	0.2092
CO <sub>2</sub>	200.8162	0.0953
H <sub>2</sub> O	5.6913	0.0027
N <sub>2</sub>	345.0911	0.1637
NH <sub>3</sub>	0.6240	0.0003
H <sub>2</sub> S	2.5377	0.0012

4 Contact between the amine solvent and raw syngas causes the chemical reactions and produces  
 5 the absorption effects. Both kinetic reactions and equilibrium reactions were adopted in the absorber  
 6 column. In this study, the associated chemical reactions include the CO<sub>2</sub>-MDEA-H<sub>2</sub>O, H<sub>2</sub>S-MDEA-  
 7 H<sub>2</sub>O and NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O systems when syngas are dissolved in MDEA solution. Reactions between  
 8 H<sub>2</sub>S and MDEA are primarily faster than CO<sub>2</sub> and MDEA owing to the selectivity of MDEA. The  
 9 governing equilibrium ion reactions with detailed equilibrium constant were given in **Table 4**. It should  
 10 be noted that the reaction equilibrium constant was calculated as a function of temperature which can  
 11 be shown in the following equation.

$$12 \quad \ln(K_{eq}) = A + \frac{B}{T} + C \ln T + DT \quad (3)$$

13 Table 4 Equilibrium reactions with adopted equilibrium constant parameters [42]

Equilibrium reactions	Equilibrium constant			
	A	B	C	D
$2H_2O \rightleftharpoons H_3O^+ + OH^-$	132.90	-13445.90	-22.48	0
$H_2O + NH_3 \rightleftharpoons NH_4^+ + OH^-$	-1.26	-3335.70	1.50	-0.04
$H_3O^+ + MDEA \rightleftharpoons MDEAH^+ + H_2O$	-9.42	-4234.98	0	0
$H_2O + H_2S \rightleftharpoons HS^- + H_3O^+$	214.58	-12995.40	-33.55	0
$H_2O + HS^- \rightleftharpoons H_3O^+ + S^{2-}$	-9.74	-8585.47	0	0
$H_2O + HCO_3^- \rightleftharpoons CO_3^{2-} + H_3O^+$	216.05	-12431.70	-35.48	0

14 On the other hand, absorption of such gases in the solution involves finite rate-controlled reaction  
 15 while equilibrium cannot be readily attained. Thus, some kinetic-controlled reactions have to be  
 16 introduced for the description of the absorption phenomenon. **Table 5** has given the chemical reactions  
 17 associated with detailed kinetic parameters and the rate law equation of kinetic reactions can be  
 18 presented by **Eq. (4)**.

$$K = k_0 \cdot \left( \frac{-E_a}{RT} \right) \quad (4)$$

Table 5. Kinetic parameters for rate-law reactions [42]

Kinetic reactions	Equilibrium constant	
	$k_0$	$E_a$ (KJ/mol)
$CO_2 + OH^- \rightarrow HCO_3^-$	4.32e+13	55.47
$HCO_3^- \rightarrow CO_2 + OH^-$	2.38e+17	123.31
$MDEA + CO_2 + H_2O \rightarrow MDEA^+ + HCO_3^-$	2.19e+06	31.10
$MDEA^+ + HCO_3^- \rightarrow MDEA + CO_2 + H_2O$	8.89e+11	64.30
$NH_3 + CO_2 + H_2O \rightarrow NH_2COO^- + H_3O^+$	1.35e+11	48.50
$NH_2COO^- + H_3O^+ \rightarrow NH_3 + CO_2 + H_2O$	4.75e+20	69.20

Radfrac non-equilibrium separation was adopted as a representative model during the absorber simulation. However, using mass and heat transfer correlations instead of the equilibrium model to predict column performance may suffer from the converge problem. In this work, consistently changing the initial values of a tear stream to reduce the converge error helps to solve the problem. Absorber to “Yes” setting and good temperature estimates should be selected and given when nonconvergent problems exist. Furthermore, the “Broyden” solver was used here for tear convergence. For the chemical absorption process, thermodynamic model ELENRTL was selected in the description of an electrolyte system. Henry components were set according to the wizard of ELENRTL choice, which included H<sub>2</sub>, CH<sub>4</sub>, CO, CO<sub>2</sub>, N<sub>2</sub>, NH<sub>3</sub> and H<sub>2</sub>S. After separating much of the H<sub>2</sub>S from the raw syngas, rich amine solution was further introduced to the regeneration column to recycle the MDEA solvent and obtain the acid gas. For the regeneration of acid gas from the liquid in the form of ion, high temperature with low pressure favours the process. It should be noted that the regeneration temperature cannot exceed the amine degradation condition approximately 137 °C [43]. For the regeneration column, only vapor-liquid equilibrium parameter was applied as the governing consideration as such kinetic reactions are not involved.

#### 2.1.4 Methanol synthesis

After the acid gas removal unit, a synthesis gas exited from the top of the absorber column with a mole fraction lower than 5 ppm H<sub>2</sub>S was ready for production of methanol [44]. Following the previous improved process design [45], an acceptable temperature of 240 °C and pressure of 75 atm was applied to the methanol synthesis. The purified syngas was first pressurized and heated before being introduced into the reactor. Then, a purge stream is used to prevent the buildup of inert gases in the reactor and control the amount of recycle stream. Detailed chemical reactions using the commercial

1 catalyst Cu/ZnO/Al<sub>2</sub>O<sub>3</sub> was shown in **Table 6**. The particle density and fixed bed voidage of the applied  
 2 catalyst were 1935kg/m<sup>3</sup> and 0.38, respectively.

3 Table 6. Methanol synthesis reactions on the Cu/ZnO/Al<sub>2</sub>O<sub>3</sub> catalyst [45]

Kinetic reactions	Reactions	Molar reaction enthalpy (25°C)
CO <sub>2</sub> hydrogenation (R1)	$CO_2 + 3H_2 \rightleftharpoons CH_3OH + H_2O$	-49 kJ/mol
Reverse water gas shift (R2)	$CO_2 + H_2 \rightleftharpoons CO + H_2O$	+41 kJ/mol
CO hydrogenation (R3)	$CO + 2H_2 \rightleftharpoons CH_3OH$	-91 kJ/mol

4 Only two reactions of the methanol synthesis were independent therefore only R1 and R2 are  
 5 implemented in the simulation tool. Owing to the governing effect of R1 thus total exothermal  
 6 performance can be seen in the reactor. It can be implied from the reaction information that to produce  
 7 more methanol, high pressure with low temperature can be favorable. The reaction kinetics on catalyst  
 8 Cu/ZnO/Al<sub>2</sub>O<sub>3</sub> have been widely studied. A common accepted kinetic description by Bussche et al [46]  
 9 was the basis of this simulation (see **Eq. (5)-(6)**). It should be noted that the original parameters should  
 10 be calculated and adjusted to directly imported into the process simulator.

$$11 \quad R1: r_{CH_3OH} = \frac{k_1 P_{CO_2} P_{H_2} - k_6 P_{H_2O} P_{CH_3OH} / P_{H_2}^3 P_{CO_2}}{(1 + k_2 P_{H_2O} / P_{H_2} + k_3 P_{H_2}^{0.5} + k_4 P_{H_2O})^3} \left[ \frac{mol}{kg_{cat} s} \right] \quad (5)$$

$$12 \quad R2: r_{RWGS} = \frac{k_5 P_{CO_2} - k_7 P_{H_2O} P_{CO} / P_{H_2} P_{CO_2}}{1 + k_2 P_{H_2O} / P_{H_2} + k_3 P_{H_2}^{0.5} + k_4 P_{H_2O}} \left[ \frac{mol}{kg_{cat} s} \right] \quad (6)$$

$$13 \quad \ln k_i = A_i + B_i / T \quad (7)$$

14 The two reactions were in vapor phase expressed in partial pressure (Pa) of involved chemicals.  
 15 All kinetic constants were described by the transformation of Arrhenius law as shown in **Eq. (7)**.  
 16 Parameters of the involved kinetic constants were summarized in **Table 7**. A multitube reactor  
 17 configuration was set in this study according to the study by Luyben [47] with the length and diameter  
 18 of 12.2 and 0.0375 meter, respectively.

19 Table 7. Kinetic constants of methanol synthesis reactions on the Cu/ZnO/Al<sub>2</sub>O<sub>3</sub> catalyst [48].

Kinetic constant	A <sub>i</sub>	B <sub>i</sub> (J/mol)
k1	-29.87	4811.2
k2	8.147	0
k3	-6.452	2068.4
k4	-34.95	14928.9
k5	4.804	-11797.5
k6	17.55	-2249.8
k7	0.1310	-7023.4

### 1 2.1.5 Methanol distillation

2 Crude methanol contained some unreacted hydrogen, CO<sub>2</sub> and water, which should be further  
3 separated to obtain high-purity methanol product. This is the aim of the methanol distillation process.  
4 The first distillation column was used to separated most of light gas while the second distillation  
5 column was applied to separated water from the methanol-water mixture. Both columns were  
6 simulated with rigorous Radfrac model. It is worth noting that by-products like methyl formate and  
7 alcohols were not included the methanol synthesis. Thus, azeotropic phenomenon did not exist in the  
8 methanol distillation process. Light gas with mainly CO<sub>2</sub> was collected in a partial condenser.  
9 Considering an effective utilization of refrigerant (-25 °C), 20 atm of the top of distillation column was  
10 selected. While the other distillation column was operated in 1 atm. Also, deign specification was  
11 conducted to obtain the separation task with 99.9% molar recovery and 99.9 molar purity of methanol.

## 12 2.2 Economic, environmental and exergy analysis

### 13 2.2.1 Economic evaluation

14 The major economic evaluation in this work is based on the research by Zhang et al [49]. The  
15 total plant cost considered in this study is comprised of two major parts namely the capital cost (*CAPC*)  
16 and the operating cost (*OPEC*). *CAPC* includes the equipment procurement (*EPC*) and installation  
17 cost (*IC*), indirect capital cost (*ICC*), working capital (*WC*) and the land cost (*LC*) for establishing the  
18 gasification (See **Eq. (8)**). *OPEC* comprises the cost of transportation cost (*TC*), feedstock cost (*FC*),  
19 utility cost (*UC*), operation and maintenance cost (*O&M*), property tax (*PT*), depreciation cost (*DC*)  
20 and general expense (*GE*) and the income tax (*IT*) (see **Eq. (9)**).

$$21 \quad CAPC = EPC + IC + ICC + WC + LC \quad (8)$$

$$22 \quad OPEC = TC + FC + UC + O\&M + PT + DC + GE + IT \quad (9)$$

23 Where *IC* is estimated by 55% of the *EPC* [50]. The *ICC* and *LC* is calculated by 123% and 6%  
24 of the total *EPC*. The total *EPC* is summed up based on each single equipment cost which is calculated  
25 using the cost correlation method as expressed by **Eq. (10)** [49].

$$26 \quad EPC = \sum_i \left( EPC_{old} \times \left( \frac{cap}{cap_{old}} \right)^n \times \frac{CEPCI}{CEPCI_{old}} \right) \quad (10)$$

27 Where *EPC<sub>old</sub>* is the base equipment cost from the previous research, besides *cap* and *cap<sub>old</sub>* are  
28 the capacity of the applied equipment now and before. Noting that the inflation is evaluated based on

1 the proportion between the chemical engineering plant cost index of different year which are denoted  
2 as  $CEPCI$  and  $CEPCI_{old}$  [51]. The averaged value of scale factor  $n$  across the whole chemical industry  
3 is about 0.6.

4 The  $ICC$  includes the cost like the official application, laboratories, supervision, canteen,  
5 auxiliary buildings, and others which are not directly to the equipment aspects. The  $WC$  is estimated  
6 by 5% of the summation of  $IC$  and  $LC$ . It can be observed from the above equations that  $EPC$  plays a  
7 crucial role in the total cost. The computation of  $EPC$  is based on a combination method of cost  
8 correlations and Aspen Process Economic Analyzer. Specialized equipment like the gasifier is  
9 simulated by different modules together. Therefore, it is better to calculate the purchased cost using  
10 cost corrections. Other common equipment like distillation column, compressor and heat exchangers  
11 can be well estimated by the economic analyzer tool. For the  $OPEC$  estimation, the  $TC$  is computed  
12 by the multiplication of transportation weight, distance, and the unit cost of transporting sewage sludge.  
13 Since there are few references about the unit transporting cost of Hong Kong and gasoline price in  
14 Hong Kong is high, three times of average unit transporting cost in Beijing was finally adopted in the  
15 estimation namely 0.3 \$/(ton\*Km) [52]. It is assumed that the average distance of sludge transportation  
16 was 37 km from West Kowloon transfer station to a sludge treatment disposal center in Lung Kwu Tan  
17 of Hong Kong (see **Fig. A2**). Combing both operating cost and capital cost, net present value ( $NPV$ )  
18 of the project can be obtained. Because of the time and inflation influence, the cash flow at present is  
19 more valuable than that in the future. Therefore, subtracting the discount cash inflow earned in the  
20 future from the initial cash outlay required for the investment provides the  $NPV$  as shown in **Eq. (11)**  
21 [49].

$$22 \quad NPV = \sum_{t=1}^{Life\ span} \frac{AR}{(1+dr)^t} - CAPC \quad (11)$$

23 Where  $AR$  is the annual revenue of a project represented by the difference of annual product  
24 income and  $OPEC$ . The discount rate is fixed at 10% for process updates along a life span of 20 years.  
25 It is considerable financially worthwhile when the  $NPV$  of a project is above zero. Moreover, internal  
26 rate of return ( $IRR$ ) provides a simpler approach to evaluate the potential of project investment for  
27 various scenarios. The  $IRR$  calculation has been shown in **Eq. (12)** and it stands for the discount rate  
28 that makes the  $NPV$  of all cash flows equal to zero. If the  $IRR$  is higher than the real discount rate, the  
29 project displays promising economic feasibility. Also, the production cost of methanol was obtained

1 by Eq. (13).

$$2 \quad \sum_{t=1}^{Life\ span} \frac{AR}{(1+IRR)^t} - CAPC = 0 \quad (12)$$

$$3 \quad Methanol\ cost = \frac{CAPC/20 + OPEC}{Amount} \quad (13)$$

4 All involved equations for economic evaluation have been summarized in **Table A3**. Nevertheless,  
5 limited economic estimation was done here and the accuracy is typically around 30% since the  
6 proposed chemical engineering process was studied at a preliminary stage [53].

### 7 2.2.2 Environmental evaluation

8 In this work, a life cycle greenhouse gas (*GHG*) emission was applied to evaluate the  
9 environmental performance of the proposed sludge treatment process with methanol production. *GHG*  
10 emissions mainly consists of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O which can be counted as the summarization of direct  
11 and indirect emission. Primary energy resources such as coal, crude oil and natura gas were consumed  
12 for the utility usage in the gasification-based methanol production process. For example, steam can be  
13 produced by burning coals which may generate much greenhouse gas and other pollutants. Herein, it  
14 is assumed that steams and required gasifier energy were supplied by the coal combustion. Meanwhile,  
15 short-distance transportation by road consumed diesel and gasoline. The energy intensity value was  
16 1362 kJ/(t.km) with an energy consumption of 68% diesel and 32% gasoline [54]. Therefore, *GHG*  
17 emissions can be calculated according to the following equations [55].

$$18 \quad CO_2 = \sum_i \sum_j E_{i,j} (D_{CO_2,j} + I_{CO_2,j}) \quad (14)$$

$$19 \quad CH_4 = \sum_i \sum_j E_{i,j} (D_{CH_4,j} + I_{CH_4,j}) \quad (15)$$

$$20 \quad NO_2 = \sum_i \sum_j E_{i,j} (D_{NO_2,j} + I_{NO_2,j}) \quad (16)$$

$$21 \quad GHG = CO_2 + 23CH_4 + 296N_2O \quad (17)$$

22 Where  $D_{CO_2,j}$ ,  $D_{CH_4,j}$  and  $D_{CO_2,j}$  are the direct emissions factors of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O for process  
23 energy  $j$  (*i.e.*, electricity, coal, diesel, and gasoline).  $I_{CO_2,j}$ ,  $I_{CH_4,j}$  and  $I_{CO_2,j}$  are the indirect emissions  
24 factors of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O for process energy.  $E_{i,j}$  is the corresponding process energy consumption  
25 of unit methanol production. The direct and indirect emission factors of the selected process energy  
26 were given in **Table A4**.

### 27 2.2.3 Exergy evaluation

28 The first law of thermodynamics indicates the amount of energy in system and surroundings are

1 conservative while it treats all the work and heat are equivalent energy without distinguishing them  
 2 from the energy grade perspective. To overcome the drawbacks in energy analysis, exergy that  
 3 indicates the maximum theoretical work extractable from heat was calculated in this work. Exergy  
 4 analysis is an advantageous method compared to conventional energy analysis that can clearly identify  
 5 the energy loss [56]. Assumptions were made before performing the exergy analysis that dead state is  
 6 set as 1 atm and 25 °C as well as kinetic exergy and potential exergy are neglected. Therefore, only  
 7 physical exergy and chemical exergy are considered in the material flow (see **Eq. (18)**). As indicated  
 8 by **Eq. (19)**, the physical exergy is determined by the enthalpy and entropy when setting 1 atm and  
 9 25 °C as the basis. The physical exergy flow rates can be well obtained using data from ASPEN Plus  
 10 and a standard spreadsheet calculation program.

$$11 \quad Ex_{total} = Ex_{ph} + Ex_{ch} \quad (18)$$

$$12 \quad Ex_{ph} = H - H_0 - T_0(S - S_0) \quad (19)$$

13 The chemical exergy refers to the maximum work of the considered stream obtainable from the  
 14 environmental state to the dead state by processes involving heat and mass transfer with the  
 15 surrounding [57]. The sewage sludge can be seen as a solid fuel, and it only considered chemical  
 16 exergy because it was introduced under the dead state. The exergy calculation of the sewage sludge  
 17 was achieved in a similar estimation of the solid fuel based on a recent work [58].

$$18 \quad Ex_{sludge} = (LHV_{sludge} + 2.442X_M) \cdot \xi + 9417X_S \quad (20)$$

$$19 \quad \xi = 1.0437 + 0.1882 \frac{H}{C} + 0.0610 \frac{O}{C} + 0.0404 \frac{N}{C} \quad (21)$$

20 The correlation factor  $\xi$  is the ratio of the chemical exergy and the lower heating value of the solid  
 21 fuel. It depends on the atomic ratios in the sewage sludge feedstocks and can be calculated according  
 22 to **Eq. (21-22)**. In addition, chemical exergy of a non-gaseous mixture and gas mixture can be defined  
 23 by the following equations respectively [56].

$$24 \quad Ex_{ch} = \sum_i n_i Ex_{stand} \quad (23)$$

$$25 \quad Ex_{ch} = \sum_i x_i Ex_{stand} + RT_0 \sum_i x_i \ln x_i \quad (24)$$

26 Where  $n_i$  is the mole flowrate of the  $i$ th component in the stream and  $x_i$  refers to the component's  
 27 molar fraction.  $R$  is gas constant that equals to 8.314 KJ/Kmol\*K.  $Ex_{stand}$  represents the standard  
 28 chemical exergy of the  $i$ th component as shown in the **Table A5**. Above all, a complete exergy balance

1 of controlled system can be formulated as **Eq. (25)**.

$$2 \quad W + \sum Ex_{Q_{in}} + \sum Ex_{in} = \sum Ex_{out} + \sum Ex_{Q_{out}} + \Delta Ex_{loss} \quad (25)$$

3 Where  $W$  is the supplied power or mechanical work, and they are equal to the exergy input.  $\sum Ex_{in}$   
 4 and  $\sum Ex_{out}$  denote the exergy flow associated with inflowing and outflowing materials, respectively.  
 5 While  $\sum Ex_{Q_{in}}$  and  $\sum Ex_{Q_{out}}$  are the utility heat source and sink, respectively. Both are rested on heat  
 6 temperature and transferred heat duty  $Q_i$ , which can be calculated by **Eq. (26)**. Exergy loss  $\Delta Ex_{loss}$  is  
 7 caused by the irreversible nature of real processes.

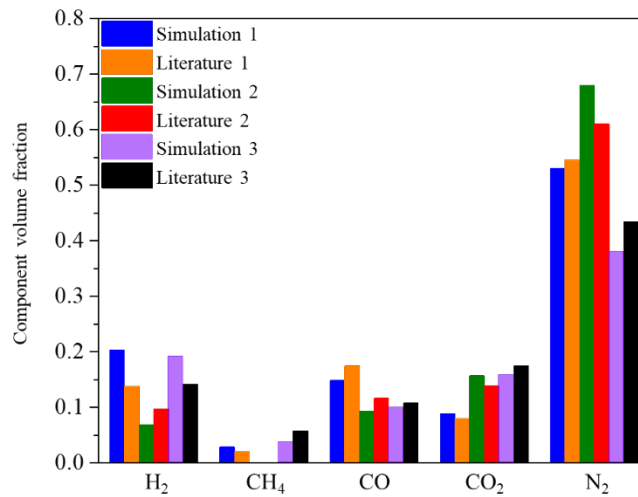
$$8 \quad Ex_Q = \sum_i \left( \left( 1 - \frac{T_0}{T_i} \right) Q_i \right) \quad (26)$$

9 Finally, exergy efficiency of a typical chemical engineering process is defined as the ratio of  
 10 output exergy and the input exergy.

$$11 \quad \eta = \frac{Ex_{product}}{Ex_{sludge} + Ex_{add}} \quad (27)$$

## 12 3. Results and discussion

### 13 3.1 Gasification results



14  
 15 Fig. 2. Gasification model validation in terms of the major syngas compositions under various conditions

16 The gasification model was verified by comparing simulation data with the experimental results  
 17 under the same operation conditions in literature 1 [28] (ER=0.28, Temperature=781 °C and Moisture  
 18 content =6.4%). In general, the gasification simulation using equilibrium model could be seen as  
 19 acceptable when the calculated average relative error (ARE) is less than 20% [35, 36]. Herein, ARE



1 was computed as 18.63% by **Eq. (28)**. It was observed from **Fig. 2** that the model prediction was  
 2 globally in good agreement with the experimental results although the N<sub>2</sub> gas has a large deviation  
 3 from the experimental results. In this case, the restricted chemical equilibrium model for the  
 4 gasification was built up with manually setting the temperature approach for R1-R5 shown in **Table 2**  
 5 equals to -150, -195, 60, -200 and 50 °C, respectively.

$$6 \quad ARE = \frac{1}{m} \sum_i^m \left| \frac{y_m - y_e}{y_e} \right| \quad (28)$$

7 Where  $y_m$  and  $y_e$  were simulation and experimental results in terms of the corresponding specified  
 8 components of major gasification gas products. Considering that the uncertainty and error of  
 9 experimental data may also affect the performance of model verification, the gasification model was  
 10 double verified by other experiments to ensure the effectiveness of the calculation results. Same  
 11 operation conditions of the literature 2 (i.e., gasifier temperature was 800 °C with feedstock sludge 6  
 12 kg/h and air flowrate 8.95 kg/h) [59] were employed in the simulation 2. Moreover, the gasification  
 13 conditions of biosolid in literature 3 [34] was 780 °C with feedstock sludge 7.05 kg/h and air flowrate  
 14 5.2 kg/h. The comparison results of simulation model were also displayed in **Fig. 2**. It could be  
 15 observed that the predicted results are in good agreement with the reported data (ARE=19.69% and  
 16 19.45%, respectively).

17 After obtaining the restricted equilibrium model parameters, we further investigate the impacts  
 18 of gasification operational factors like mass moisture content, air equivalent ratio ( $ER$ ) and gasification  
 19 temperature. Moisture content can affect the equilibrium of gasification reactions and higher  
 20 temperature required more external energy addition.  $ER$  is the ratio between the actual air molar  
 21 quantity and the theoretical air supply for complete combustion. For a given sludge feedstock with  
 22 specified ultimate results, the assumed composition can be written as  $CH_aO_bN_cS_d$  then  $ER$  can be  
 23 calculated by the following equation.

$$24 \quad ER = \frac{m}{(1 + a/4 - b/2 + c + d) \times 4.76} \quad (29)$$

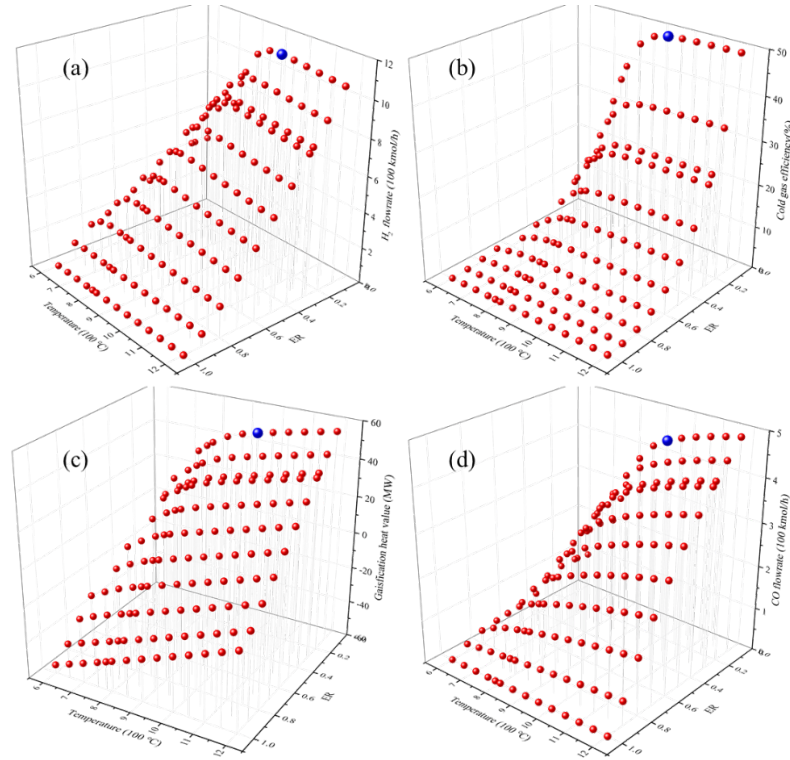
25 Where  $m$  is the actual molar flowrate of air supplement. In the sensitivity analysis, the influences  
 26 of above-mentioned factors on the H<sub>2</sub> production, syngas composition, lower heating value ( $LHV$ ) of  
 27 syngas and cold gas efficiency ( $CGE$ ) were investigated. The  $LHV$  (MJ/Nm<sup>3</sup>) of product gas depends  
 28 on the volume percentage of CO, H<sub>2</sub> and CH<sub>4</sub> in the raw syngas and can be obtained through **Eq. (30)**

1 [60]. The *CGE* is defined as the ratio of the flow of energy in the syngas and the energy contained  
 2 within the feedstock sludge (See **Eq. (31)**).

$$3 \quad LHV_{gas} = 35.88x_{CH_4} + 12.63x_{CO} + 10.78x_{H_2} \quad (30)$$

$$4 \quad CGE(\%) = \frac{v_{gas} \times LHV_{syngas}}{M_{sludge} \times LHV_{sludge}} \quad (31)$$

5 Where  $x$  is the volume fraction of gas products after the gasification reaction.  $v_{gas}$  is the total  
 6 volume (Nm<sup>3</sup>) of CO, H<sub>2</sub> and CH<sub>4</sub>. The value of  $LHV_{sludge}$  is 25.63 MJ/kg according to the study [32].



7  
 8 Fig. 3. The influence of gasification temperature and air flowrate on (a) the H<sub>2</sub> flowrate; (b) the CGE  
 9 performance; (c) gasification energy consumption; and (d) CO flowrate.

10 Three major variables namely the gasification temperature, *ER* and the moisture content were  
 11 changed to investigate the metrics variation of produced H<sub>2</sub>, CO, the CGE and gasification energy  
 12 consumption. The gasification temperature was ranged from 600 to 1200 °C. *ER* value was changed  
 13 from 0.1 to 1.0. Moreover, moisture content was adjusted from 0 to 60%. Three variables were  
 14 simultaneously changed in the simulation and the optimal operation conditions in terms of the largest  
 15 amount of hydrogen flowrate were *ER*=0.1, 950 °C and 60 % moisture content which was shown in  
 16 blue balls in the following **Fig. 3** and **4**. It can be observed from **Fig. 3** that blue balls in each subplot  
 17 were not always the optimal results. Nevertheless, the amount of H<sub>2</sub> gas was set as the objective of

1 sensitivity analysis because the hydrogen gas plays an important role in the methanol synthesis. As  
2 shown in **Fig. 3(a)**, with the decreasing air flowrate, more hydrogen can be produced from the gasifier.  
3 This may be caused by the water gas shift reaction under the condition of more carbon monoxide that  
4 produced by partial oxidation of carbon solid. Also, **Fig. 3(a)** reveals that higher temperature is  
5 beneficial for hydrogen production. *CGE* is a key metrics for evaluating the performance of energy  
6 conversion during the gasification. Generally, if more combustible components like H<sub>2</sub>, CH<sub>4</sub> and CO  
7 was generated then much higher *CGE* will be in display. The trend of *CGE* resembles that of H<sub>2</sub> and  
8 CO flowrate. This may be ascribed to the dominated amount of H<sub>2</sub> and CO gases. The gasification heat  
9 value equals to the heat summarization of drying, pyrolysis and gasification subsections. It was implied  
10 from **Fig. 3(c)** that the complete combustion with enough air will release much heat and thus the total  
11 gasification heat duty shown a negative value.

12 **Fig. 4** was dedicated to demonstrating the influence of moisture content on four indicators H<sub>2</sub>  
13 flowrate, CO flowrate, *CGE* and energy consumption. Higher moisture content favours in the hydrogen  
14 production while poses a converse effect on the CO generation. In addition, it needs more energy  
15 consumption during the gasification section because of the endothermic nature of water gas reaction  
16 and Boudouard reaction in the gasification process. As shown in **Fig. 4**, the blue ball cannot represent  
17 the best *CGE* and CO flowrate, nor the minimized total energy consumption. However, the blue ball  
18 stands for the maximum H<sub>2</sub> flowrate which is a key factor influencing the methanol synthesis. To some  
19 extent, the sewage sludge gasification provides a renewable approach for the H<sub>2</sub> production since H<sub>2</sub>  
20 was mainly obtained through the natural gas reforming or electrolytic water [61].

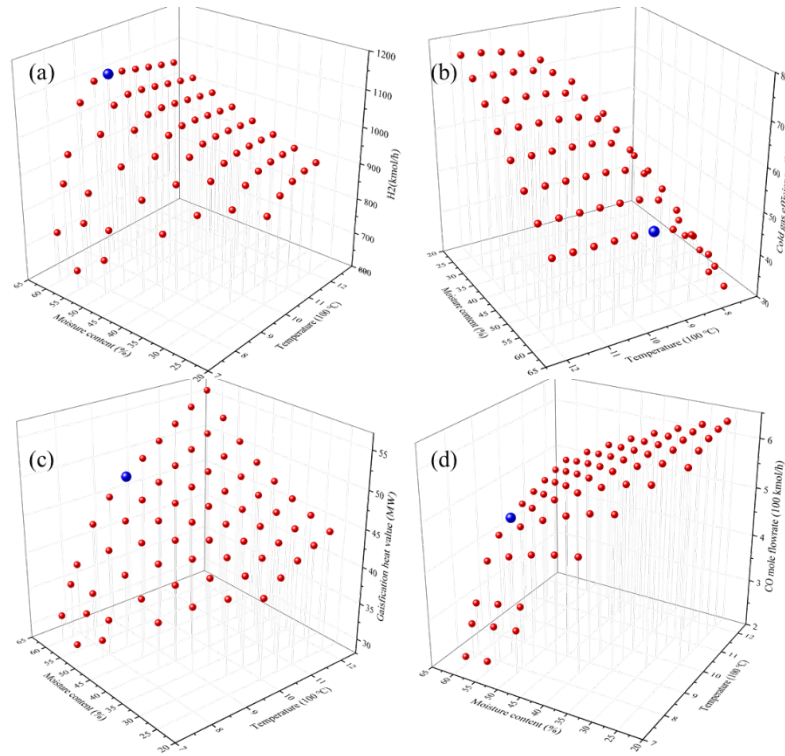


Fig. 4. The influence of gasification temperature and moisture content on (a) the H<sub>2</sub> flowrate; (b) the CGE performance; (c) gasification energy consumption; and (d) CO flowrate.

### 3.2 Power generation analysis

To recover heat energy from the high-temperature stream, steam cycle power generations were implemented in this work. Some essential factors namely the water flowrate, cold stream outlet temperature, discharged pressures of pump and steam turbine greatly influence the power generation. Firstly, the flowrate of water and superheat degree of steam were adjusted to achieve the largest value of heat duty. It represented the maximum heat recovery from the crude syngas stream. Then, the discharged pressures of the pump and steam turbine was adjusted to obtain a maximum brake work of steam turbine. It must be noted that some results from the sensitivity analysis of the power generation shown the temperature cross in the heat-exchanger simulation, thus these points should be deleted. The sensitivity analysis for two power generation subprocesses were given in **Fig. 5** and **6**, respectively. The first steam cycle process was used to recover heat energy from raw syngas. The cycled water flowrate was adjusted from 1800 to 1850 kmol/h and the superheat degree of outlet steam was varied in a range from 1 to 15 °C. In addition, the discharge pressure of pump and steam turbine were set in ranges 40-54 atm and 0.92-1.20 atm, respectively.

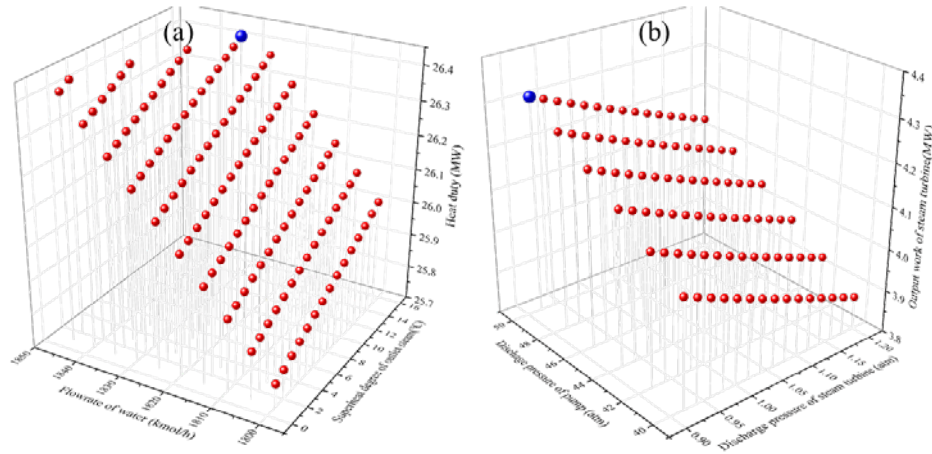


Fig. 5. (a) The influence of water flowrate and superheat degree of steam on the heat duty of recycled energy; (b) The influence of discharge pressures of pump and steam turbine on the produced work

As shown in **Fig. 5(a)**, the blue ball represents the optimal condition associated with a water flowrate of 1830 kmol/h and a superheat degree of steam of 14 °C. Actually, larger water flowrate and steam temperature will recovery more heat energy from raw syngas. However, it is limited by the heat exchanger operating with no temperature cross. Then, maximum power generation was given under the largest pressure difference between the pump (*i.e.*, 50 atm) and steam turbine (*i.e.*, 0.9 atm).

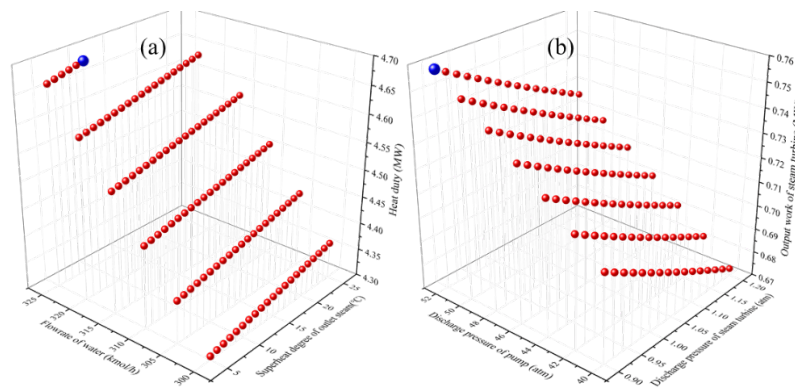


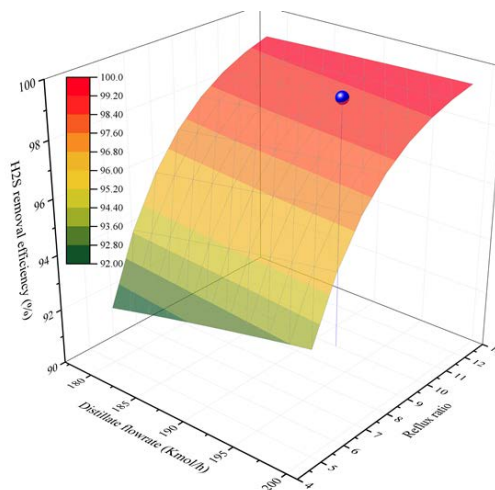
Fig. 6. (a) The influence of water flowrate and superheat degree of steam on the heat duty of recycled energy; (b) The influence of discharge pressures of pump and steam turbine on the produced work

Before the raw syngas was introduced into the absorption column, it should be compressed and cooled to a suitable condition for efficient removal of acid gas. Therefore, another steam cycle process can be used to recover the heat energy. The water flowrate in the cycle varied from 300 to 325 kmol/h and the superheat degree of outlet steam was changed from 5 to 25 °C. Furthermore, the discharge pressure of pump and steam turbine were set in ranges 40-52 atm and 0.90-1.20 atm, respectively. The sensitivity analysis results were shown in **Fig. 6**. It reveals that the optimal conditions for maximum

1 recovered heat were 320 kmol/h water flowrate and superheat degree of 10 °C. They are less compared  
2 to that in the first steam process. The obtained pressures of pump and steam turbine in the second  
3 power generation section were 52 atm and 0.9 atm.

### 4 3.3 Absorption process

5 The major aim of this section is to remove most of H<sub>2</sub>S component from the syngas and obtain  
6 the clean gas for methanol synthesis. And a low temperature and high pressure is good for the gas-  
7 liquid solubility as well as the absorption. The raw syngas was sent into the top of absorption column  
8 at 10 atm and 35 °C. The design specification of absorption column was provided in **Table A6**. In  
9 general, two key factors namely circulation rate and the concentration of MDEA solution greatly affect  
10 the H<sub>2</sub>S removal efficiency. To optimize the operation parameters, the ratio of circulation rate to feed  
11 raw syngas (L/G) was varied from 0.1 to 1.0 under the fixed 5 mol% MDEA concentration. Then, the  
12 MDEA concentration was changed from 1 mol% to 10 mol% targeting on investigating the H<sub>2</sub>S  
13 removal efficiency. The sensitivity analysis results have been shown in **Fig. A3**. Results indicated that  
14 increased L/G ratio and MDEA concentration is benefit for removing H<sub>2</sub>S substance. It should be noted  
15 that the H<sub>2</sub>S in feedstock only account for 0.12 mol% of the raw syngas. Therefore, it can be readily  
16 achieved to obtain 99.99% removal when L/G larger than 0.5 and MDEA concentration larger than 5  
17 mol%. In this regard, the L/G and MDEA concentration were selected as 0.5 and 5 mol%.



18 Fig. 7. The effect of reflux ratio and distillate flowrate on the H<sub>2</sub>S removal efficiency of the stripper column  
19 After the absorption column, the bottom stream that contains H<sub>2</sub>S component with 2.5591 kmol/h  
20 was sent into the regeneration stripper column. The regeneration column was maintained at lower  
21 pressure and high temperature due to advantages in distillation. The pressure of stripper column is 1  
22 atm. And a heat exchanger was used to raise the feed temperature to 85°C. This preheating  
23

1 configuration can decrease the reboiler duty of regeneration column. Different from the absorber  
 2 column, a vapor-liquid equilibrium model was used in the stripper. Considering the energy  
 3 consumption, we set the molar recovery of H<sub>2</sub>S as 99% which means at least 2.5335 kmol/h H<sub>2</sub>S should  
 4 be collected from the top of stripper column. Then, an analysis was conducted with respect to the  
 5 influence of reflux ratio and distillate flowrate on the H<sub>2</sub>S removal efficiency. As shown in **Fig. 7**, both  
 6 reflux ratio and distillate flowrate do a favour in increasing the H<sub>2</sub>S removal efficiency. However,  
 7 higher efficiency means more reboiler duty for the separation. Moderate 99% of H<sub>2</sub>S recovery was  
 8 selected (see blue ball in **Fig. 7** and the corresponding condition was given associated with distilled  
 9 flowrate of 192 kmol/h and reflux ratio of 10. Then, the total number of stages was 15 which was  
 10 chosen based on the convergence requirement of simulations. **Fig. 8** has shown the detailed  
 11 information of the absorption process. Molar flowrate of related ion components was not displayed  
 12 individually. Apparent flowrate was given in **Fig. 8** to better demonstrate the electrolyte system.

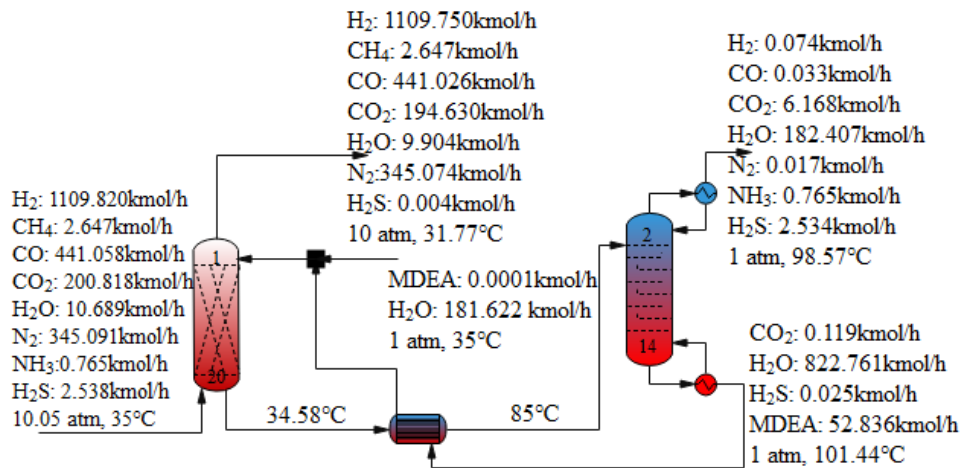


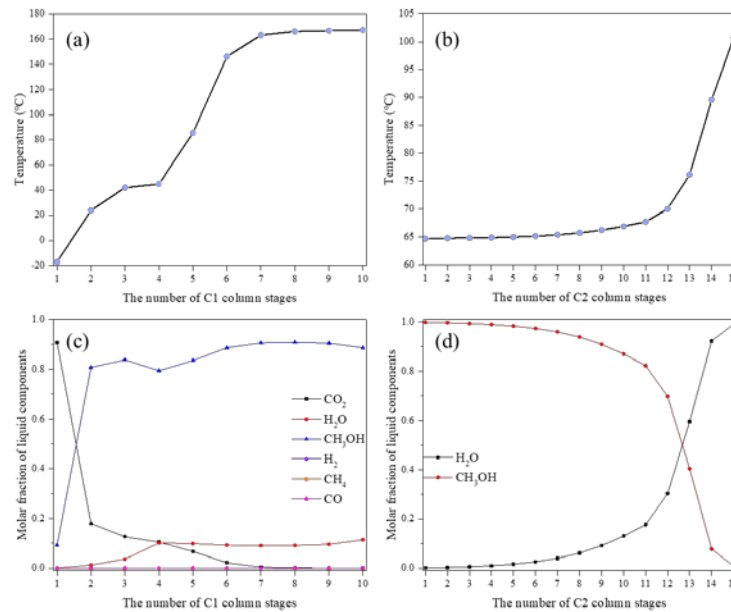
Fig. 8. The absorption process flowsheet with apparent molar flowrate

### 3.4 Methanol process

16 The methanol synthesis was a mature technology, and the simulation was finished according to  
 17 the methodology demonstration in **Section 2.1**. The pressure and temperature in methanol synthesis  
 18 reactor were chosen as 75 atm and 240 °C [45, 62]. RK-SOAVE equation of state was applied in  
 19 calculating related physical chemistry parameters. A fraction of recycled gas during the synthesis was  
 20 purged to avoid gas cumulation. Obtained crude methanol with detailed compositions was given in  
 21 **Table A7**. It is evident that major impurities come from the water and other light gas. It is required to  
 22 make a purification and thus two equilibrium-based distillation columns were applied. The first  
 23 distillation column aimed at separating most light gas from the mixture. With varying the distillate

1 flowrate, the molar recovery of CO<sub>2</sub> can be achieved above 99.9%. Meanwhile, the sensitivity analysis  
 2 was conducted to investigate the influence of reflux ratio on the final reboiler duty of the first  
 3 distillation column (see **Fig. A4**). The optimal reflux ratio is found to be 0.1 associated with a distillate  
 4 flowrate of 88.913 kmol/h. Because a refrigerant utility with -25 °C temperature was used for partial  
 5 condensation of low-temperature light gas. The column pressure was adjusted at 20 atm to meet the  
 6 heat-transfer requirement.

7 Second distillation column operated at the atmospheric pressure was used to separate water and  
 8 methanol. Operating at 1 atm can guarantee the cooling water useful for the condenser. Different from  
 9 the analysis of the first distillation column, the methanol purity should be considered as well. As such  
 10 two design specifications were set to control the methanol recovery (99.9%) and its purity (99.9%) by  
 11 changing the distillate flowrate and reflux ratio, respectively. Eventually, the obtained distillate rate  
 12 and reflux ratio were 490.076 kmol/h and 1.109. In addition, Suitable number of stages and feed stages  
 13 of two distillation columns were set according to the temperature profiles of two columns. As shown  
 14 in **Fig. 9 (a)** and **(b)**, each plate in both distillation columns caused separation effects with clear  
 15 temperature variations. It was evident from **Fig. 9 (c)** that few light gasses existed in the bottom of  
 16 distillation column C1 and the feed stage is 4<sup>th</sup> tray. Since there is no azeotrope the distillation column  
 17 C2 can well obtained methanol product with 99.9 mol% purity. The column profiles indicated both  
 18 distillation columns were well optimized.



19

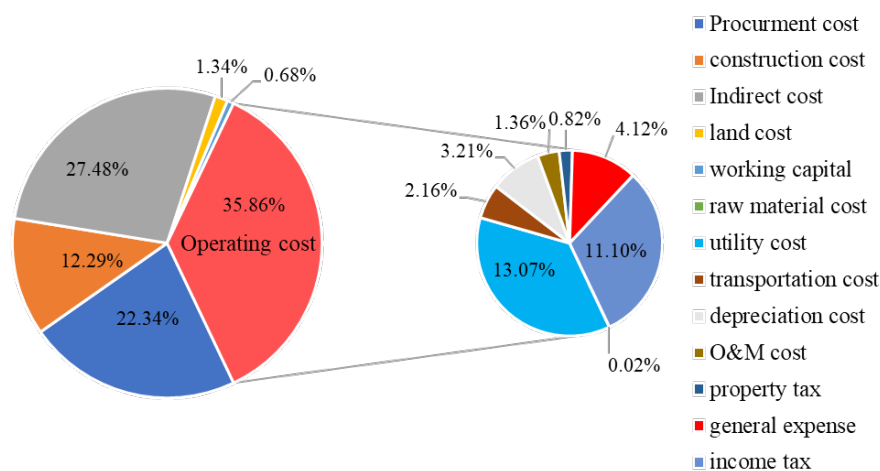
20

Fig. 9. (a) (b) Temperature profiles and (c) (d) major liquid composition in distillation column C1 and C2



## 1 3.5 Economic, environmental and exergy analysis

### 2 3.3.1 Economic results



3  
4 Fig. 10. Economic cost breakdown for the gasification process

5 Through the sensitivity analysis of key variables in different subsections, detailed streams mass  
6 flowrates have been obtained and summarized in **Appendix B**. Followed by a sustainability analysis  
7 in terms of different aspects, whether if the STM plant is promising should be answered. Detailed  
8 estimation cost of the STM process was shown in **Table A8**. The economic cost breakdown for the  
9 *CAPC* and *OPEC* of the initial gasification was shown in **Fig. 10**. It was shown that *OPEC* accounts  
10 for only 35.86% of the total cost and the remain 64.14% of the total cost was the *CAPC*. This was in  
11 accord with the practical chemical engineering plant. Large amount of initial investment was the  
12 equipment related cost (*i.e.*, *CAPC*). Indirect cost (25.24 million \$) which includes miscellaneous  
13 overhead, field services, labour benefits and et al dominated the *CAPC* followed by the equipment  
14 procurement (131.73 million \$) and construction cost (45.89 million \$). The right pie chart displayed  
15 the breakdown of *OPEC* and it revealed that utility consumption and income tax was the major cost  
16 contributor which takes up 13.07% and 11.10%, respectively. The price of involved chemicals and  
17 utility energy has been given in **Table A9**. Notably, pure nitrogen gas was separated from a pressure-  
18 swing adsorption equipment and the referred price of nitrogen gas product was 0.00123/m<sup>3</sup> at a  
19 condition of 15 Mpa and 25 °C.

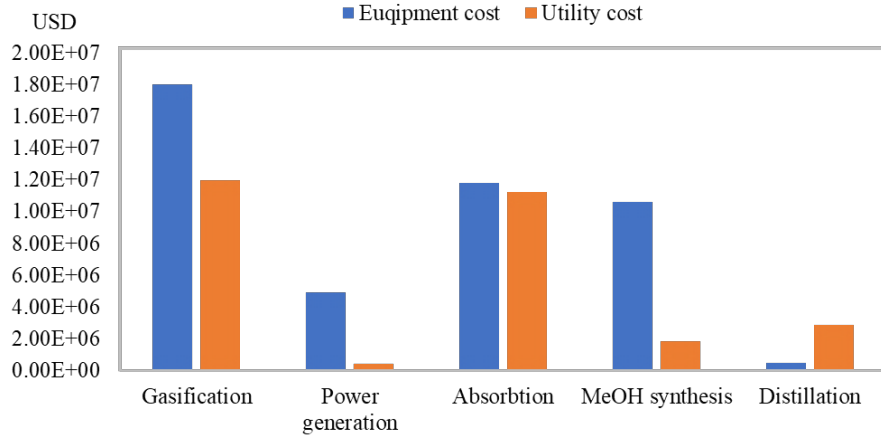


Fig. 11. Equipment cost and utility cost of subsections of the proposed conversion process

To make a clearer comparison of subsections in the process, we then calculated the equipment and utility cost in different subprocesses. As shown in **Fig. 11**, there is no doubt that the gasifier system consumed the largest energy to maintain the required gasification state of high temperature condition. The equipment procurement cost in the gasification section was 18.04 million\$ and the gasification consumed a high-temperature fuel cost for 12 million\$. It was implied from the calculation that the gasification system in the proposed system still has the potential to be further optimized. Absorption process is second largest expensive in terms of the equipment cost (11.79 million\$) because it included a costly compressor. Moreover, the procurement cost (10.60 million\$) of methanol synthesis has a similar value of that of absorption process. In terms of the methanol distillation, the process did consume high-pressure steam in the reboiler and low-temperature refrigerant in the condenser. The utility usage is much lower compared with that in the gasification system.

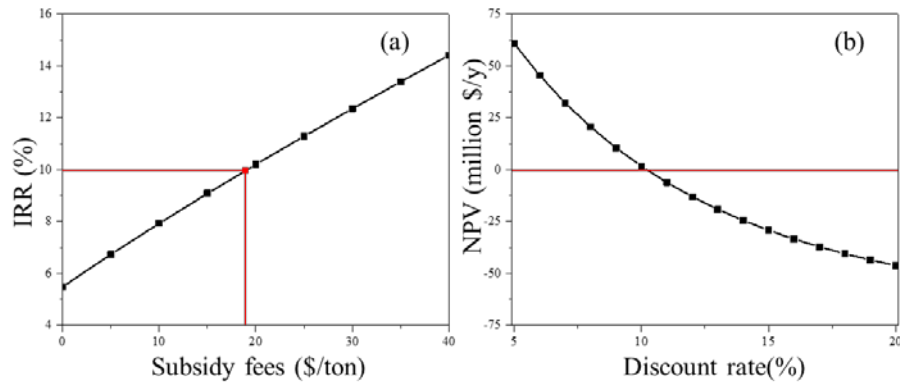


Fig. 12. (a) the effect of subsidy level on the final IRR of the project and (b) the effect of discount rate on the NPV of the project in 20 years life span

The calculated cost of sewage sludge gasification-based treatment is 39.58 \$/ton which is lower than the conventional sludge treatment by combustion in China mainland (approximately 61.54-76.92

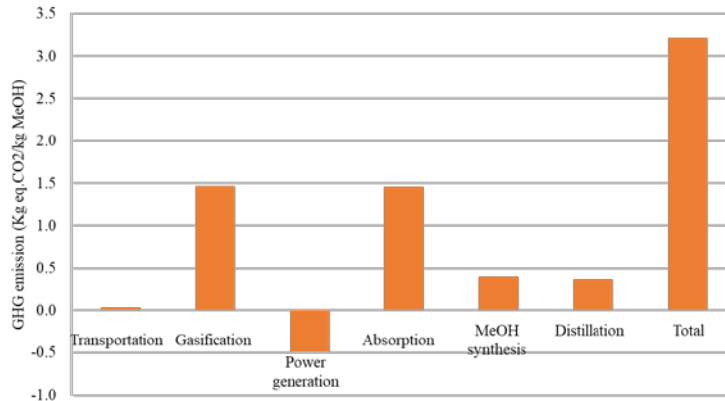
1 \$/ton [63]). The methanol production cost by proposed STM process was 579.62 \$/ton, and this value  
2 is in accord with that in the recent summary report by International Renewable Energy Agency (*i.e.*,  
3 400-800 \$/ton) [64]. Nevertheless, as shown in **Table A8**, it is hard for the proposed STM plant to  
4 achieve beneficial economics in the case of no subsidies provided by local government as the *NPV*  
5 value is negative based on assumed 10% discount rate. Meanwhile, calculated *IRR* value is only 5.48%  
6 representing the critical value of discount rate that makes *NPV* equal to zero. To better understand the  
7 importance of public financial support, **Fig. 12 (a)** have shown the influence of subsidy fees on the  
8 final *IRR*. Notably, the analysis was conducted under the assumption of no price fluctuations of  
9 methanol and by-product nitrogen. It was indicated from **Fig. 12 (a)** that an increased subsidy support  
10 does a favour in the economic benefits. Generally, 10% of *IRR* is used as a good value to evaluate the  
11 economic feasibility of a project when considering the currency inflation. In light of this, a subsidy of  
12 20 \$/ton could be suitable for the STM plant implementation.

13 On the other hand, the price of methanol produced in this way is relatively high. Therefore, the  
14 effect of methanol price fluctuation on the economic indicator *IRR* should be further conducted to  
15 identify the feasibility of STM process. To evaluate this influence, the sensitivity analysis method was  
16 performed by giving a variation of  $\pm 20\%$  in the methanol price. **Fig. A5** has shown the analysis results  
17 and the basic methanol price is set as 450 \$/ton. When the methanol price is more than 461.68 \$/ton  
18 under the circumstance of no subsidy fees, the project was economically feasible. This is in accordance  
19 with the negative *NPV* value that shown in **Table A8**. After 20 \$/ton support is given, the project will  
20 be economically doable when methanol price is more than 403.88 \$/ton. With the increased methanol  
21 price, the project becomes more profitable.

22 **Fig. 12 (b)** has reflected the influence of discount rate on the *NPV* when giving a subsidy of  
23 20\$/ton. In this case, if the actual discount rate is much larger than 10%, total cash flow in 20 years  
24 will be negative which means the project conduction would be not doable. The discount rate can be  
25 referred as the interest rate charged to commercial banks and other financial institutions for short-term  
26 business loans. According to the statistics of business bank in Hong Kong [65], average interest rate  
27 of 6.5% was shown in the past 40 years. Therefore, it was indicated from **Fig. 12 (b)** that a subsidy of  
28 20\$/ton associated with 6.5% discount rate was enough for achieving the positive *NPV*. In summary,  
29 the STM project with sludge disposal capacity of 1200 ton/day is promising in the economic aspect at  
30 a cost of public support. At the initial stage of a new greener technology, this kind of public support is

1 an effective method to promote a greener and circular economy.

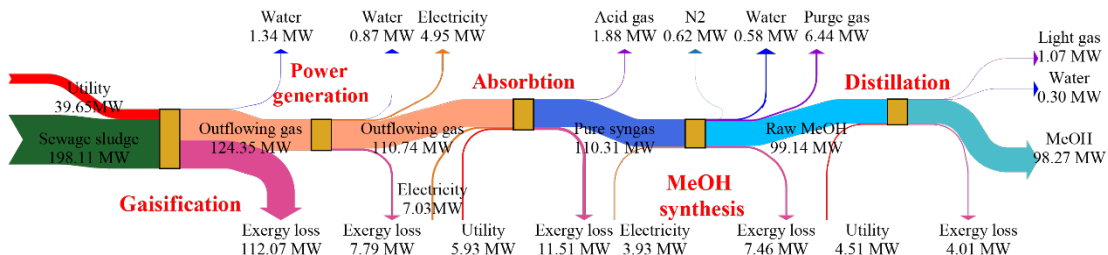
2 3.3.3 Environmental results



3  
4 Fig. 13. Life cycle GHG emissions of subprocesses for the proposed methanol production

5 LCA environmental analysis of the proposed process was performed using the simulation results  
6 according to **Eq. (14-17)** and corresponding emission factors. The function unit was 1kg produced  
7 methanol. Eventually, **Fig. 13** has given the GHG emissions of subsections for the STM process. The  
8 total GHG emission of the methanol production from the treatment of sludge was 3.21kg eq.CO<sub>2</sub>.  
9 Relatively large GHG emissions were shown in the gasification and absorption sections. The GHG  
10 emission of gasification and absorption subsections were 1.47 and 1.45 kg eq.CO<sub>2</sub>, respectively. It is  
11 evident that power generation section can mitigate the carbon emission by 0.50 kg eq.CO<sub>2</sub>. The net  
12 carbon reduction of power generation is limited. To some extent, gasification and absorption process  
13 accounts for large carbon emissions which implies the bottleneck of further improvement. To reduce  
14 the environmental pollution and conform to the low-carbon manufacturing, future research is necessary  
15 to explore the energy integration network to reduce the GHG emissions caused by the utility  
16 consumption of gasification units.

17 3.3.2 Exergy results



18  
19 Fig. 14. The exergy flow of the STM treatment process

20 According to a sludge to methanol study which focus on the energy efficiency [66], the overall  
21 exergy efficiency is about 53%. For the proposed processes, the exergy streams of the system have

1 been shown in **Fig. 14**. Only methanol was set as the product and the calculated exergy efficiency was  
2 37.92%. The total exergy input is about 259.16 MW which including the utility input and sewage  
3 sludge exergy. During the gasification-based utilization process, the air gasification system has the  
4 largest exergy destruction (*i.e.*, 112.07 MW) followed by the absorption system (*i.e.*, 11.51 MW). The  
5 exergy loss during the methanol synthesis section is 7.46 MW and it accounts for 5.22% of the total  
6 exergy destruction while during gasification accounts for 78.46%. The detailed distribution of exergy  
7 destruction was given in **Fig. A6**. A reason for the largest exergy loss in gasification was the high-  
8 temperature condition associated with high irreversibility of chemical reactions. In summary, further  
9 improvement can be carried out by focusing on increasing the energy efficiency of gasification and  
10 absorption sections through the process system engineering technology.

#### 11 4. Conclusion

12 A sludge-to-methanol process was conceptually designed by considering the case in Hong Kong.  
13 Based on the detailed simulation in Aspen Plus, sensitivity analysis of key operational parameters was  
14 conducted such as gasification temperature, ER and moisture content after the validation of the  
15 restricted Gibbs equilibrium gasification model. Economic results revealed the IRR of the STM plant  
16 is 5.48% under the scenario of no subsidy support. The sludge disposal cost and methanol production  
17 cost were 39.58 and 579.62\$/ton, respectively. An averaged IRR value of 10% is appropriate for the  
18 economic feasible thus a subsidy of 20\$/ton sludge should be given to obtain a positive NPV. In  
19 addition, the total lifecycle GHG emission is 3.21 kg eq.CO<sub>2</sub>/kg methanol. GHG emissions of the  
20 gasification section is the largest because of the high temperature condition should be maintained by  
21 consuming lots of utility coals. The overall exergy efficiency of STM is 37.92% and relatively  
22 dominated proportion of exergy loss was shown in the gasification and absorption, which indicated  
23 both subsections still have room for further improvement. In terms of the practical implication, this  
24 work can give some guidelines for the gasification utilization of sludge in Hong Kong. It is implied  
25 from the exergy analysis that how to improve the energy efficiency of sludge gasification and the  
26 syngas cleaning should be the future focus which may involve more pilot-scale experiments and energy  
27 integration network design. Economic analysis indicated the commercial start-up of this plant was not  
28 easy. Public subsidy must be given to the plants to support the technical retrofit and future innovation.  
29 Detailed environmental impacts comparison was not studied in this research while we provided a GHG

1 emission value for the reference. In the near future, a detailed comparative life cycle research should  
2 be conducted to show how good of the STM process is when compared to conventional methanol  
3 production processes. During the environmental comparison analysis, it is also considerable to involve  
4 the carbon tax concept.

5 It should be noted that the model construction of the sewage sludge to methanol conversion was  
6 finished based on the restricted thermodynamic equilibrium model. This Aspen Plus model conducted  
7 the complex gasification process simulation in a simpler way as the small sections (e.g., pyrolysis,  
8 gasification and fully combustion) in the gasification were tested and adjusted individually. Therefore,  
9 it does not need to cover all the chemical reactions in complex reacting systems. These advantages  
10 have contributed to the wide application of equilibrium models in the design and evaluation of solid  
11 coal gasification especially happened in the large-scale downdraft gasifier. However, the weakness of  
12 the applied model is obvious. Some assumptions like that tar were not considered and the heat loss  
13 was neglected were firstly given to simplify the gasification-based methanol production models. Those  
14 assumptions may not practically be accurate to reproduce all the experimental results. Further  
15 improvement should be made in the model accuracy by considering artificial neural network models  
16 and detailed kinetic models.

## 17 Declaration of Competing Interest

18 The authors declare that they have no known competing financial interests or personal  
19 relationships that could have appeared to influence the work reported in this paper.

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## 25 Appendix

26 The Supplementary material is available free of charge via the Internet.

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