



Carbon-negative co-production of methanol and activated carbon from bagasse pyrolysis, physical activation, chemical looping, and methanol synthesis

Guangcan Su^{a,b}, Nurin Wahidah Mohd Zulkifli^{a,b,1,*}, Li Liu^{c,1,*}, Hwai Chyuan Ong^d, Shaliza Ibrahim^e, Kai Ling Yu^f, Yifan Wei^g, Feng Bin^{h,i}

^a Department of Mechanical Engineering, Faculty of Engineering, University of Malaya, Kuala Lumpur 50603, Malaysia

^b Centre for Energy Sciences, University of Malaya, Kuala Lumpur 50603, Malaysia

^c School of Environment, Tsinghua University, Beijing 100084, China

^d School of Engineering and Technology, Sunway University, Jalan Universiti, Bandar Sunway, 47500 Petaling Jaya, Selangor, Malaysia

^e Institute of Ocean and Earth Sciences (IOES), University of Malaya, Kuala Lumpur 50603, Malaysia

^f Tan Sri Leo Moggie Distinguished Chair in Energy Informatics, Institute of Informatics and Computing in Energy (IICE), Universiti Tenaga Nasional (UNITEN), Malaysia

^g School of Mechanical Engineering, Jiangsu University, Zhenjiang 212013, China

^h State Key Laboratory of High-Temperature Gas Dynamics, Institute of Mechanics, Chinese Academy of Sciences, Beijing 100190, China

ⁱ School of Engineering Science, University of Chinese Academy of Sciences, Beijing 100049, China

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ABSTRACT

Methanol is regarded as an important chemical precursor in the chemical industry and has huge potential to replace gasoline and diesel as vehicle fuel. Biomass to methanol is a sustainable and green production method, but its economic and environmental viability is contingent on production technologies and geographic context. This study proposed a carbon-negative methanol production method that integrated four modules of bagasse pyrolysis, physical activation, chemical looping, and methanol synthesis in the context of China. Three scenarios, including co-production of methanol and biochar, co-production of methanol and activated carbon, and co-production of methanol and activated carbon with extra hydrogen, were put forward and simulated in Aspen Plus. An evaluation system was established to quantitatively assess the carbon and energy efficiencies and economic and environmental benefits of the three scenarios. The results suggested that the addition of hydrogen effectively increased the methanol yield in Scenario 3, leading to high carbon and energy efficiencies. Scenarios 1 and 2 exhibited better economic and environmental performance with low payback periods of 6.53 and 5.80 years and low global warming potentials of -1631.18 and -710.28 kg CO₂-eq/t methanol. However, Scenario 3 would be economically and environmentally feasible by decreasing hydrogen production costs and implementing green hydrogen production methods in the foreseeable future. This study provides a viable approach for sustainable methanol production in China, thereby aligning with the current imperative of achieving carbon neutrality.

Abbreviations: AC, Activated carbon; BTM, Biomass-to-methanol; CCS, Carbon capture and storage; CTM, Coal-to-methanol; EC, Equipment cost; EOW, Electrolysis of water; GWP, Global warming potential; IC, Installation cost; IRR, Internal rate of return; LCA, Life-cycle assessment; MEA, Monoethanolamine; NPV, Net present value; NTM, Natural gas-to-methanol; OMC, Operation and maintenance costs; PBP, Payback period; PPC, Process plant cost; PSA, Pressure swing adsorption; S-I cycle, Sulphur-iodine cycle; SMR, Steam methane reforming; TCC, Total capital cost; TEA, Techno-economic analysis; TPC, Total plant cost; NPCC, National Plan for Climate Change; WCI, Working capital investment.

* Corresponding authors at: Department of Mechanical Engineering, Faculty of Engineering, University of Malaya, Kuala Lumpur 50603, Malaysia (N. W. M. Zulkifli).

E-mail address: nurinmz@um.edu.my (N.W.M. Zulkifli).

¹ The two authors have the same contribution to this study.

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1. Introduction

The methanol industry is seen as the hub of a flourishing chemical industry network. As an important industrial chemical, methanol is a precursor for the synthesis of various chemicals, such as dimethyl ether, formaldehyde, anisole, methyl benzoate, and acetic acid. Furthermore, methanol is also a clean and high-quality liquid fuel with high combustion efficiency and possesses huge potential to replace gasoline and diesel as vehicle fuel [1]. As the global leader in methanol production, the total production of methanol in China was 78.16 million tons in 2021, accounting for 59% of the global production capacity [2]. However, the methanol industry in China is an energy-intensive industry that heavily relies on coal, and the coal-to-methanol (CTM) route accounts for approximately 65% of methanol production, which differs from the global prevailing natural gas-to-methanol (NTM) process [3]. The dominant CTM technology causes severe CO₂ emissions; the carbon footprint of the CTM technology chain is about 2970 kg CO₂-eq/t methanol [4]. The biomass-to-methanol (BTM) technology offers a promising approach to mitigate global warming from the source and produces methanol in a green and sustainable way. China is the largest producer of biomass resources in the world, with an annual output of 3.49 billion tons [5]. Bagasse is one of the agricultural wastes with the largest output in the world. China is the main sugarcane-producing country in the world, and the annual sugarcane production is approximately 70 million tons, producing about 20 million tons of bagasse [6]. The rational development and utilization of bagasse are of great significance to protect the environment and promote the sustainable development of energy in China.

Meanwhile, the substitution from coal to biomass can relieve CO₂ emissions by decoupling methanol synthesis from fossil resources. Bai et al. [7] built a solar-biomass gasification polygeneration system to co-produce electricity and methanol, and the results suggested that the system could yield 54800 tons of methanol and 50.85 GW-h of electricity with a high exergy efficiency of 51.89%. Yang et al. [8] proposed a BTM process with efficient entrained flow gasification. The BTM process demonstrated a low biomass consumption of only 1.99 t/t methanol while achieving a high methanol yield of 18.5 mol/kg with a high exergy efficiency of 70%.

However, the BTM route is confronted with many problems. Gasification is an effective technology for converting biomass into syngas, but it requires a significant amount of energy and a controlled supply of oxygen, air, or steam [9]. The biomass pyrolysis is introduced to address these challenges. The pyrolysis process can transform biomass into syngas, char, and bio-oil at lower temperatures (400–700 °C) under anoxic environment without oxygen, air, or steam [10]. Solid product can be isolated as a byproduct, and syngas can be converted into hydrogen-enriched syngas through steam reforming [11]. Olaleye et al. [12] found that pyrolysis/gasification can produce more hydrogen than gasification alone, and that the hydrogen yield can be enhanced by using steam reforming or partial oxidation of bio-oil. Pyrolysis has advantages over gasification in terms of energy consumption and scale flexibility [13].

Moreover, biomass-derived syngas is still deficient in hydrogen, and syngas conditioning for CO₂ capture is significant in methanol synthesis. Monoethanolamine (MEA) absorption is a mature and effective CO₂ capture technology that has been commercially available and extensively used in various industries for over half a century. However, this technology still faces many issues, such as high energy consumption, corrosiveness, and MEA degradation [14]. Moreover, pressure swing adsorption (PSA) is a new technique for the removal of CO₂. PSA is based on selectively or preferentially adsorbing CO₂ onto the solid adsorbent under relatively high pressure by contacting the hot gas with the solid adsorbent in the packed column. Then, the adsorbed CO₂ is desorbed from the solid by reducing the partial pressure of the gas phase in the column so that the adsorbent can be reused. Nevertheless, low purity, complex design, and high energy consumption hinder its further

promotion [15]. The chemical looping process is relatively novel and possesses great application potential in pyrolysis, gasification, reforming, and combustion of biomass, which is a cyclic process performed using a looping material as a set of sub-reactions to capture or release CO₂ [16]. It employs solid carrier materials to collect CO₂ and avoid the energy-intensive gas–liquid separation step, resulting in high energy efficiency. Meanwhile, the utilization of cheap CO₂ carriers, such as CaO and Fe₂O₃, makes it economically competitive with traditional processes [17]. This technology can be scaled up to an industrial scale, allowing the large-scale production of fuels and chemicals [18]. These strengths make the technology have enormous application potential.

Techno-economic analysis (TEA) is a methodology used to evaluate the technical and economic feasibility of a process or technology, which can inform decision-makers investing in a particular technology or process and improve its economic viability [19]. Many scholars have employed the TEA tool to assess the economic feasibility of novel methanol synthesis routes. Ye et al. [20] performed and compared the TEA of a poly-generation system (methanol and electricity) with integrated gasification combined cycle, and they found that the poly-generation system achieved better economic performance with a low payback period (PBP) of 4.4 years. Meanwhile, the environmental performance of bioenergy from biomass is an important factor in determining the feasibility of the project under economically viable conditions. Life-cycle assessment (LCA) is extensively employed to assess the environmental impacts of a product, service, or process over its whole lifecycle, which has been utilized in many cases of methanol synthesis by various researchers [21]. For instance, Meunier et al. [22] reported a CO₂-to-methanol technology roadmap and estimated its environmental effect through LCA. About 1300 kg CO₂-eq was released to produce 1 ton of methanol, far less than the traditional technologies.

However, the introduction of the chemical looping process in methanol synthesis is relatively underexplored in the existing literature. Meanwhile, maximizing the utilization of biomass resources through the co-production of methanol and biochar/activated carbon (AC) has not been reported before. Consequently, this study proposed a novel method for the co-production of methanol and biochar/AC from bagasse pyrolysis, physical activation, chemical looping, and methanol synthesis, aiming to replace traditional fossil-based conversion pathways with more sustainable and greener routes. Three scenarios, including co-production of methanol and biochar, co-production of methanol and AC, and co-production of methanol and AC with extra hydrogen, were simulated, analyzed, and compared. The whole process was analyzed and optimized for methanol synthesis by adjusting various conditions. Carbon and energy efficiencies of the three scenarios were evaluated and computed. The economic and environmental performance of the three scenarios were determined through TEA and LCA. This study provides important insights into the viability of this BTM approach and highlights its potential for reducing the carbon footprint of the methanol industry.

2. Methodology

2.1. Experimental method

The elemental and ultimate properties of bagasse is a crucial indicator for determining its pyrolysis characteristics. Table S1 summarizes the properties of bagasse. The bagasse sample was crushed and sifted through sieves to obtain various particles with a size range from 0.075 to 0.15 mm and dried at 60 °C for 24 h. As in our previous study, the pyrolysis experiment was conducted in a tube furnace at 700 °C for 30 mins [23].

2.2. Process design

Fig. S1 depicts the detailed Aspen Plus flowsheet of methanol synthesis from bagasse, and the whole system consisted of pyrolysis and steam reforming unit, activation unit, chemical looping unit, and

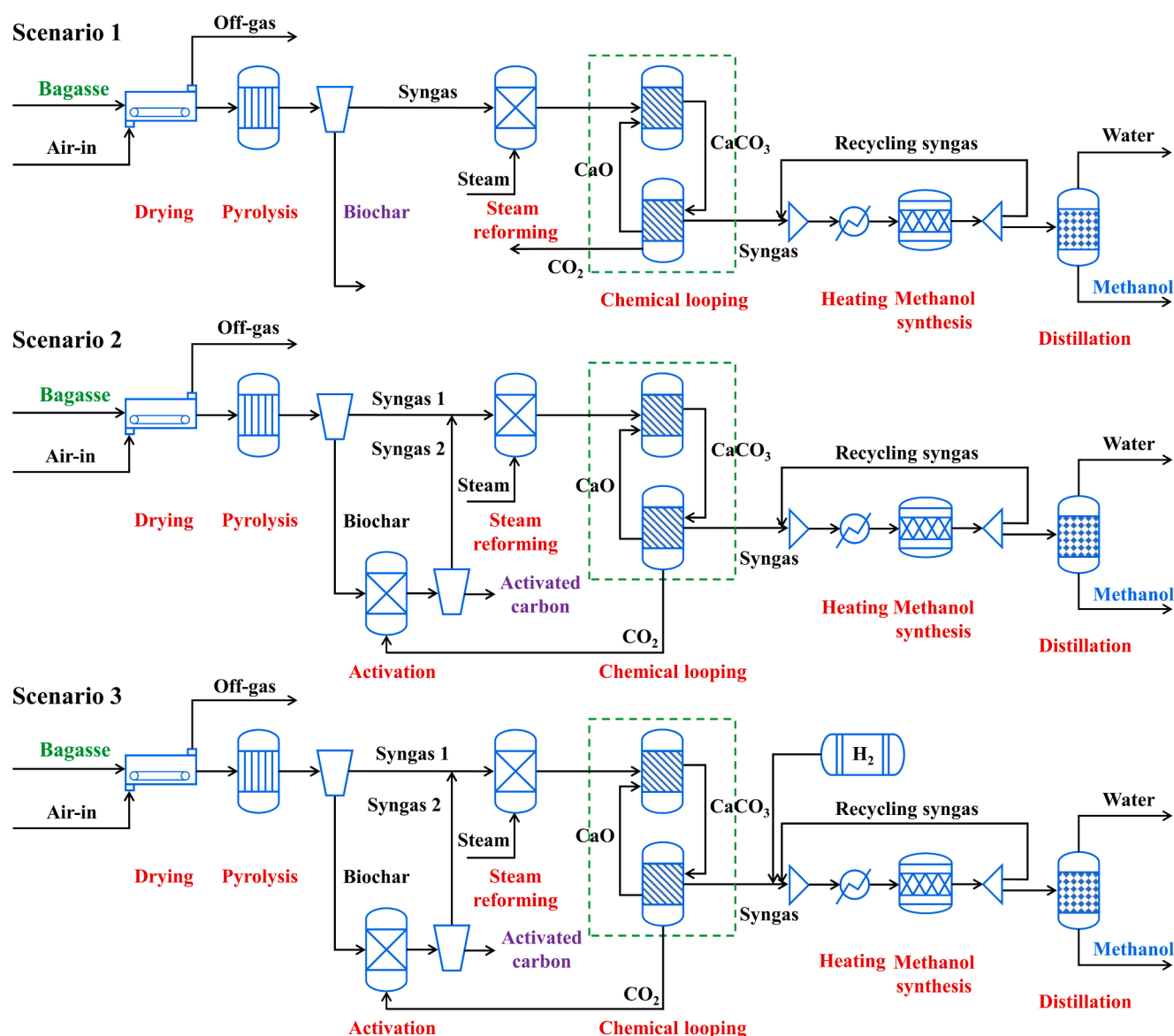


Fig. 1. Schematic flowchart of the three scenarios.

methanol synthesis and distillation unit. Table S2 lists the description of process blocks in Aspen Plus. In the drying process, the moisture content of bagasse was decreased to a reasonable value. Subsequently, dry bagasse was fed into the pyrolysis process, solid and gaseous products were separated, and the solid product was sent to the activation process. The resulting syngas was mixed with the gaseous products from pyrolysis and sent to the steam reforming process, and then transferred to the chemical looping unit. At last, the adjusted syngas was delivered into the methanol synthesis and distillation unit to produce methanol. As presented in Fig. 1, three scenarios are proposed based on different production conditions. Scenario 1 is the co-production of methanol and biochar without the activation unit, Scenario 2 is the co-production of methanol and AC, and Scenario 3 uses additional hydrogen to increase methanol yield.

The Aspen Plus simulation treated bagasse as a nonconventional component with its enthalpy and density defined as HCOALGEN and DCOALIG, respectively. Ash was also defined as a nonconventional component; the stream class was designated as CONVEN, and global streaming was utilized to model unconventional materials. The Peng-Robinson equation of state was selected as the base property method due to its suitability for hydrocarbon-processing and gas-processing

applications. The system throughput was set at 10 t/h. Assumptions made in the study included: (1) All reactions were under isothermal conditions and steady state, (2) all reactions in the modules reached equilibrium instantaneously and maintained steady-state reactions, (3) ash was considered a non-reactive material and did not participate in the reaction, (4) the inlet stream temperature and pressure were 25 °C and 1 atm, respectively, (5) there was no heat or temperature loss in the pipeline or reactor, (6) tar and macromolecular substances generated in pyrolysis were not included, and (7) bagasse was assumed to participate completely in the reaction.

2.2.1. Pyrolysis, activation, and steam reforming processes

In the pyrolysis process, bagasse was converted into conventional components (N_2 , H_2 , O_2 , CO , CO_2 , CH_4 , H_2O , C , and ash) at 700 °C in an RYield reactor based on the experiment data from our previous study [24]. Afterward, the solid product was delivered to the activation process through an RGibbs module, and CO_2 derived from the chemical looping process acted as an activator and induced the activation reaction, as presented in Eq (1). The resulting syngas was mixed with the gaseous products from bagasse pyrolysis. Syngas underwent reforming reactions to generate H_2 and CO according to thermodynamic

Table 1
Parameters values of kinetic models [28].

Model constant	Coefficient	Unit	Value
k_1	A_1	–	1.07
	B_1	J/mol	40,000
k_2	A_2	–	3453.38
	B_2	J/mol	–
k_3	A_3	–	0.499
	B_3	J/mol	17,197
k_4	A_4	–	6.62×10^{-11}
	B_4	J/mol	124,119
k_5	A_5	–	1.22×10^{10}
	B_5	J/mol	–98084

Table 2
Techno-economic assumptions.

Economic parameters	Unit	Value
Assumptions		
Plant lifetime	Year	20
Building cycle	Year	1
Production in the preparation period	Year	The production rate is 0.7, 0.8, and 0.9 in the 2nd, 3rd, and 4th years, respectively.
Operation time	h/year	8500
Throughput	t/h	10
Income tax	%	25
Sale tax	%	27
Bank interest ^a	%	6.4
Input-fixed costs		
Equipment cost (EC)	USD	Aspen Plus
Installation cost (IC)	USD	Aspen Plus
Process plant cost (PPC)	USD	EC + IC
Total plant cost (TPC)	USD	130% of PPC
Total capital cost (TCC)	USD	110% of TPC
Working capital investment (WCI)	USD	20% of TCC
Operation and maintenance costs (OMC) ^b	USD	4% of TCC
Input-variable costs		
Bagasse ^c	USD/t	37.79
Hydrogen ^d	USD/t	4840
Cu/ZnO/Al ₂ O ₃ catalyst ^e	USD/t	10,000 (5 years)
Catalyst dosage ^f	t	1.73
CaO ^g	USD/t	92.86
CaO dosage	t	5
Transportation fee ^h	USD/t	Fixed cost + Variable cost × Distance
Fixed cost	USD/t	0.85
Variable cost	USD/(km·t)	0.12
Distance	km	50
Input-utility costs		
Electricity ⁱ	USD/kWh	0.0775
Natural gas ^j	USD/MMBtu	7
Water for industry ^k	USD/t	0.70
Output		
Methanol ^l	USD/t	402.88
Biochar ^m	USD/t	400 (400 to 500)
AC ⁿ	USD/t	1138
Techno-economic indicators		
NPV	Million USD	$NPV = \sum_{n=0}^N \frac{PV_n}{(1+i)^n} \times (1+i)^{-n}$
IRR	%	$0 = \sum_{n=0}^N \frac{PV_n}{(1+IRR)^n}$
PBP	Year	PBP = Years with negtive NPV+ NPV /PV

^a The bank interest and sale and income taxes are obtained from the study [24].

^b Assumptions are acquired from the literature [32].

^c The bagasse price is obtained from the quotation, and the USD to CNY exchange rate was 6.910 [36].

^d The hydrogen price in China in 2022 is obtained from the Shanghai Environment Energy Exchange [37].

^e Catalyst cost is obtained from the paper [38].

^f Catalyst dosage is based on the paper [25].

^g CaO price is acquired from the paper [39].

^h Transportation fee is obtained from the paper [33].

ⁱ The electricity price is acquired from the paper [23].

^j The natural gas cost is obtained from the paper [40].

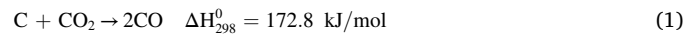
^k The water price is obtained from the report [41].

^l The methanol price is acquired from the paper [34].

^m The biochar price is obtained from the report [42].

ⁿ The AC price is acquired from the literature [23].

equilibrium in an RGibbs reactor. Subsequently, syngas was fed into the chemical looping process for purification and adjustment.



2.2.2. Chemical looping process

Syngas from bagasse pyrolysis and biochar activation was delivered into the chemical looping process. Syngas and steam were fed into an RGibbs reactor to convert H₂O and CO into H₂ and CO₂ and realize the capture of CO₂ in syngas through the carbonation reaction, as shown in Eq (2) to Eq (4). Then, gas and solid products were separated by a Cyclone Separator, and the obtained syngas was delivered into a Flash to remove water for subsequent methanol synthesis. CaCO₃ was sent into

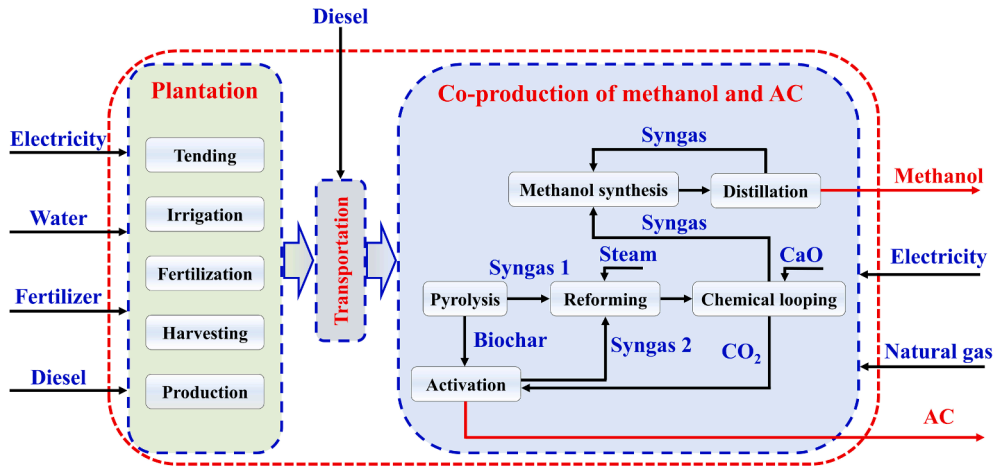
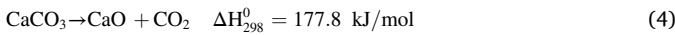
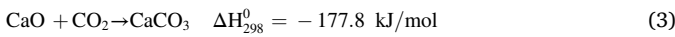
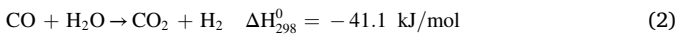


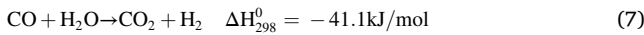
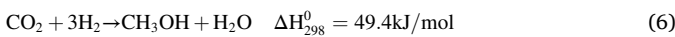
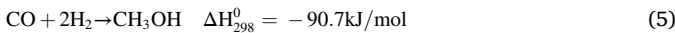
Fig. 2. System boundary for LCA analysis.

an RGibbs reactor to transform CaCO_3 into CaO and CO_2 through the calcination reaction, and CaO was recycled for the next round of CO_2 separation. Then, the generated CO_2 was delivered into the physical activation process as an activator.



2.2.3. Methanol synthesis and distillation processes

In the methanol synthesis and distillation processes, the operating conditions for methanol synthesis were set as 220.5 °C and 50 bars. Syngas was heated to the target temperature through a heater, then fed into the multi-tubular RPlug reactor and compressed to the target pressure to produce methanol based on the following reactions:



The multi-tube catalytic reactor has a distinctive feature, consisting of 810 tubes that are 12 m long and 0.06 m in diameter and loaded with 865 kg of $\text{Cu}/\text{ZnO}/\text{Al}_2\text{O}_3$. The properties of the catalyst are listed in Table S3 [25]. The reactor effluent underwent phase separation to separate the reaction products from the unreacted gases, which were then recycled and pressurized before being mixed with the reactor feed. Methanol was separated from water in a distillation column using the RadFrac module based on the rigorous equilibrium stage model [26]. Then, products were separated into syngas and methanol through a Flash, and the left syngas was recycled into the multi-tubular RPlug reactor to reproduce methanol.

The kinetic model was on the basis of the study from Bussche and Froment [27] with the readjusted parameters from Mignard and Pritchard [28] (Eq. (8) and (9)), where the temperature was expressed in K and the pressure in bar. The kinetic constants were based on the Arrhenius law (Eq. (10)), and its parameters are summarized in Table 1. Thermodynamic equilibrium constants were obtained from the research [29] (Eq. (11) and (12)). However, those models cannot be implemented directly in software as Aspen Plus only supports certain kinetic equation types. Therefore, kinetic models and related parameters were readjusted to allow the reactor simulation in Aspen Plus. The pressure drop in the reactor was computed through the Ergun equation, which had been implemented in the software.

$$r_{\text{CH}_3\text{OH}} = \frac{k_1 P_{\text{CO}_2} P_{\text{H}_2} \left(1 - \frac{1}{K_{\text{eq}}} \frac{P_{\text{H}_2\text{O}} P_{\text{CH}_3\text{OH}}}{P_{\text{H}_2} P_{\text{CO}_2}}\right)}{\left(1 + k_2 \frac{P_{\text{H}_2\text{O}}}{P_{\text{H}_2}} + k_3 P_{\text{H}_2}^{0.5} + k_4 P_{\text{H}_2\text{O}}\right)^3} \left[\frac{\text{mol}}{\text{kg}_{\text{cat}} \text{s}}\right] \quad (8)$$

Reverse water-gas shift reaction.

$$r_{\text{RWGS}} = \frac{k_5 P_{\text{CO}_2} \left(1 - K_{\text{eq}2} \frac{P_{\text{H}_2\text{O}} P_{\text{CO}}}{P_{\text{CO}_2} P_{\text{H}_2}}\right)}{\left(1 + k_2 \frac{P_{\text{H}_2\text{O}}}{P_{\text{H}_2}} + k_3 P_{\text{H}_2}^{0.5} + k_4 P_{\text{H}_2\text{O}}\right)^3} \left[\frac{\text{mol}}{\text{kg}_{\text{cat}} \text{s}}\right] \quad (9)$$

$$k_i = A_i \exp\left(\frac{B_i}{RT}\right) \quad (10)$$

$$\log_{10} K_{\text{eq}1} = \frac{3066}{T} - 10.592 \quad (11)$$

$$\log_{10} \frac{1}{K_{\text{eq}2}} = -\frac{2073}{T} + 2.02 \quad (12)$$

2.3. Carbon and energy efficiencies

In the background of global warming, carbon efficiency is an important indicator to evaluate the environmental friendliness of the project. The carbon element in raw materials is distributed in the product in a certain proportion after the reaction, and the calculation of carbon efficiency is based on the conservation of carbon element mass before and after the reaction, that is, the ratio of the carbon element mass in the product to the carbon element mass in the raw material. One of the current obstacles in biomass utilization is incomplete biomass utilization, which means that carbon in biomass is lost during the utilization process or directly converted into CO_2 . Therefore, improving the carbon efficiency of biomass can not only increase the economic efficiency of biomass utilization but also enhance the carbon reduction capability of biomass. As shown in Eq (13), carbon efficiency is the ratio of carbon mass in the product to that in the raw material [30].

$$\text{Carbon efficiency} = \frac{\text{Carbon in the product}}{\text{Carbon in the feedstock}} \quad (13)$$

The present scenario entails the utilization of energy-intensive processes, such as pyrolysis and activation, which consume significant amounts of energy. Consequently, the recovery of energy from end products and internal applications assumes critical importance in realizing economic effectiveness and environmental benefits. Therefore, energy efficiency analysis is employed to evaluate the self-sustainability of the three scenarios. As displayed in Eq (14), energy efficiency assessment is calculated based on two key components: energy

Table 3
Life-cycle inventory data.

Items	Category	Unit	Value	
Plantation (for the production 1 ton bagasse) ^a	Phosphorous fertilizer	kg	8.00	
	Potassium fertilizer	kg	9.00	
	Nitrogen fertilizer	kg	2.40	
	Water	m ³	243.00	
	Electricity	kWh	6.60	
	Diesel	lit	0.80	
CO₂ emission coefficient	Fertilizer ^b	kg CO ₂ -eq/kg	0.05	
	Groundwater ^c	kg CO ₂ -eq/m ³	0.156	
	Electricity ^d	kg CO ₂ -eq/kWh	0.549	
	Diesel ^e	kg CO ₂ -eq/lit	2.68	
CO₂ emission coefficient of hydrogen preparation	SMR ^f	kg CO ₂ -eq/kg	10.00	
	SMR with CCS ^g	kg CO ₂ -eq/kg	5.61	
	S-I cycle ^g	kg CO ₂ -eq/kg	9.16	
	S-I cycle with nuclear ^h	kg CO ₂ -eq/kg	0.41	
	S-I cycle with solar ^h	kg CO ₂ -eq/kg	1.02	
	EOW ^g	kg CO ₂ -eq/kg	27.60	
	EOW with solar ⁱ	kg CO ₂ -eq/kg	3.20	
	EOW with wind ^j	kg CO ₂ -eq/kg	0.88	
	EOW with nuclear ^j	kg CO ₂ -eq/kg	0.76	
	Biomass ^j	kg CO ₂ -eq/kg	2.60	
	Biomass with CCS ^j	kg CO ₂ -eq/kg	-14.58	
Transportation^k		kg CO ₂ -eq/(t-km)	0.17	
Project				
	Pyrolysis and steam reforming unit	Electricity	kg CO ₂ -eq/kWh	0.549
	Activation unit	Natural gas ^l	kg CO ₂ -eq/MMBtu	53.07
	Chemical looping unit	Electricity	kg CO ₂ -eq/kWh	0.549
Methanol synthesis unit	Electricity	kg CO ₂ -eq/kWh	0.549	

^a The inventory for sugarcane plantation is obtained from the literature [45].

^b The carbon footprint of fertilizer is obtained from the report [48].

^c The carbon footprint of groundwater is obtained from the study [49].

^d The CO₂ emission of the electricity in China is obtained from the report [47].

^e The carbon footprint of diesel utilization is obtained from the report [50].

^f The CO₂ emissions of SMR is acquired from the literature [51].

^g The CO₂ emission of the S-I cycle and EOW is acquired from the paper [52].

^h The CO₂ emission of the S-I cycle with nuclear power or solar energy is acquired from the literature [53].

ⁱ The CO₂ emission of EOW with solar is acquired from the literature [54].

^j The CO₂ emission of the EOW is acquired from the literature [55].

^k The CO₂ emission of transportation is acquired from the literature [46].

^l The data is acquired from the EIA [56].

consumption and energy recovery. Energy consumption refers to the energy required for the entire process of methanol and biochar/AC production. In contrast, energy recovery refers to the energy in the final product [31].

$$\text{Energy efficiency} = \frac{\text{Energy in the product}}{\text{Energy in the feedstock} + \text{Energy in utilities}} \quad (14)$$

2.4. Techno-economic analysis

Profitability is a key factor in determining the economic viability of a project, and TEA is commonly used to assess the commercial viability of a process. The TEA of bagasse-to-methanol and biochar/AC involves developing a flowsheet in Aspen Plus to calculate capital and operation costs based on the mass and heat balances. Table 2 summarizes detailed economic assumptions in this work and economic computed results from Aspen Plus. Aspen Plus is a powerful tool for economic analysis, and the equipment and installation costs can be obtained from Aspen Plus

Economic Analyzer. Other economic indicators are typically estimated using literature sources [32]. The plant life and the construction duration are 20 years and 1 year, respectively. The plant is assumed to operate at 8500 h per year and has a bagasse processing capacity of 10 t/h. Electricity is utilized to supply energy for pyrolysis and steam reforming, chemical looping, and methanol synthesis and distillation unit, and the price of electricity is 0.0775 USD/kWh in China. Natural gas is used to support the operation of the activation process. The transportation logistics for the project involve the utilization of an 8-t medium diesel truck to transport feedstocks over 50 km (round-trip). The transportation cost is determined using a linear function that accounts for both fixed cost and variable cost factors. This approach enables a more comprehensive analysis of transportation costs, which is a critical component of the overall economic feasibility assessment for the project [33]. The price of methanol was collected from the previous literature [34].

The economic viability of the project is evaluated using three key techno-economic indicators: net present value (NPV), PBP, and internal rate of return (IRR). These indicators provide a comprehensive assessment of the financial performance of the project over its lifetime. In addition, the discount rate used in the analysis is based on the prevailing rate in China, which is estimated to be approximately 3% [35]. Furthermore, sensitivity analysis is employed to evaluate the impact of various factors on the economic performance of the project and to assess its overall robustness, which provides insight into the vulnerability of the project to external factors and enables a more comprehensive assessment of its long-term financial viability. The sensitivity analysis on PBP conducted in this study focuses on seven key variables: transport distance, electricity, feedstock cost, biochar/AC price, methanol price, TCC, and hydrogen cost, enabling a more comprehensive assessment of the economic performance of the project under various conditions and factors.

2.5. Life-cycle assessment

LCA is a widely recognized methodology for evaluating the environmental impact of biomass conversion and utilization processes [43]. In the study, the environmental impact of the three scenarios was assessed using a cradle-to-gate approach, following the ISO14040:2006 standard, enabling a comprehensive assessment of the environmental performance of the project, from its initial stages of development through the production of the final product [44]. As depicted in Fig. 2, the system was divided into three distinct components: sugarcane plantation, transportation, and conversion process.

Table 3 lists the life-cycle inventory data of the project. The systematic and comprehensive research on the inventory for sugarcane plantations, including tending, fertilization, irrigation, harvesting, and collection, was conducted in the study [45]. 8-t medium diesel truck is used for transportation, and its emission coefficient is 0.17 kg CO₂-eq/(t-km) [46]. The consumed energy in the conversion process is provided by electricity and natural gas; the CO₂ emission coefficients of electricity and natural gas are 0.549 kg CO₂-eq/kWh and 53.07 kg CO₂-eq/MMBtu, respectively [47]. The CO₂ emission coefficient of hydrogen preparation has a high dependence on the production process and energy source. Traditional production methods, such as steam methane reforming (SMR), sulphur-iodine cycle (S-I cycle), and electrolysis of water (EOW), have high CO₂ emission coefficients. However, the application of novel technologies, including carbon capture and storage (CCS) and renewable energy sources, including solar, nuclear, and wind, can remarkably mitigate CO₂ emissions and even make the production process a carbon-negative process.

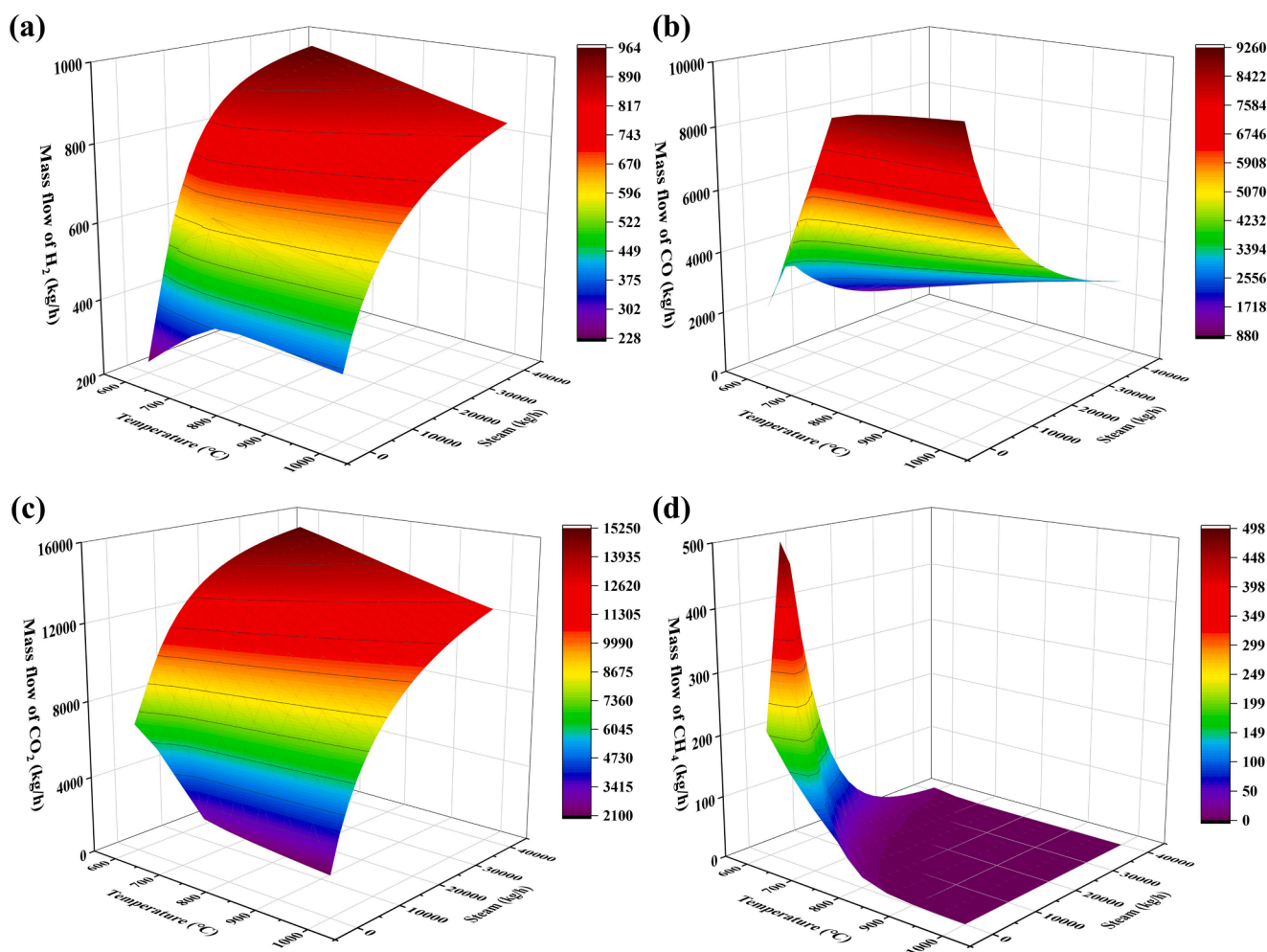


Fig. 3. Effects of temperature and steam in GAS-1 on the yield of (a) H_2 , (b) CO, (c) CO_2 , and (d) CH_4 in Scenario 2.

3. Results and discussion

3.1. Process analysis and optimization

The selection of reforming conditions, including temperatures and steam/biomass, is crucial to maximize syngas production and minimize undesirable byproducts. Fig. 3 illustrates the effect of various temperatures and steam rates on the yield of different gaseous products in GAS-1 (Conditions in GAS-2: 700 °C, 5000 kg/h) in Scenario 2. The increment of the steam flow rate had a positive impact on H_2 yield at all simulated temperatures, as a high steam rate is favorable for water–gas shift and methane reforming reactions. Nevertheless, the increase in temperature under various steam flow rates affected the H_2 yield in different ways. At a low steam flow rate of 2500 kg/h, the H_2 yield increased with growing temperature, while at a high steam flow rate, it decreased with the increasing temperature. The CO production first increased to 6799.67 kg/h and then decreased with the increase in the steam flow rate at a low temperature of 700 °C. However, the CO yield declined with the growth of the steam flow rate at high temperatures. Meanwhile, the increment in temperature exhibited a promotional effect on the formation of CO. As for CO_2 , it presented a downward trend with the rising temperature at all steam flow rates and an upward trend with the increasing steam flow rate at all temperatures. In contrast, CH_4 declined with the rise in temperature and steam flow rate. Overall, the rising steam flow rate increased the yield of H_2 and CO_2 and reduced the yield of CO and CH_4 due to the water–gas shift and methane reforming reactions, where the steam reacts with CO and CH_4 to form CO_2 and H_2 [57]. Meanwhile, the

increasing temperature raised the yield of CO and H_2 , as high temperatures can increase the degree of primary fragmentation of biomass macromolecules and promote several endothermic reactions, including the Boudouard and reforming reactions [58].

Fig. S2 depicts the effect of various conditions in GAS-1 (Conditions in GAS-2: 700 °C, 5000 kg/h) and GAS-2 (Conditions in GAS-1: 700 °C, 5000 kg/h) on the methanol yield in Scenario 2. In GAS-1, the steam rate played a leading role in methanol production. The increment in temperature increased the methanol yield at a low steam rate, whereas the rise in temperature had a limited influence on the methanol yield at high steam rates. However, the methanol production raised sharply as the steam rate increased from 2500 to 5000 kg/h, and then the methanol production increased slowly with the rising steam rate. As for the GAS-2, the temperature had a dominant influence on the methanol yield, and 700 °C was the optimal temperature for methanol production. Consequently, the condition for GAS-1 and GAS-2 was set at 700 °C, 5000 kg/h (steam/biomass = 0.5), and 700 °C, 5000 kg/h (steam/biomass = 0.5), respectively, after a tradeoff between methanol yield and energy consumption. Fig. S3 displays the effect of input CaO in the chemical looping process on the methanol and AC yield. The increment in the CaO amount continuously increased the methanol yield and decreased the AC yield. The increase of CaO amount during the chemical looping process generated more CO_2 activators for biochar activation. The increase in the amount of CO_2 allowed more carbon in the biochar to be converted into CO during biochar activation. Subsequently, more CO entered the steam reforming process and reacted with H_2O to generate H_2 , thereby increasing methanol yield. From the perspective of ensuring

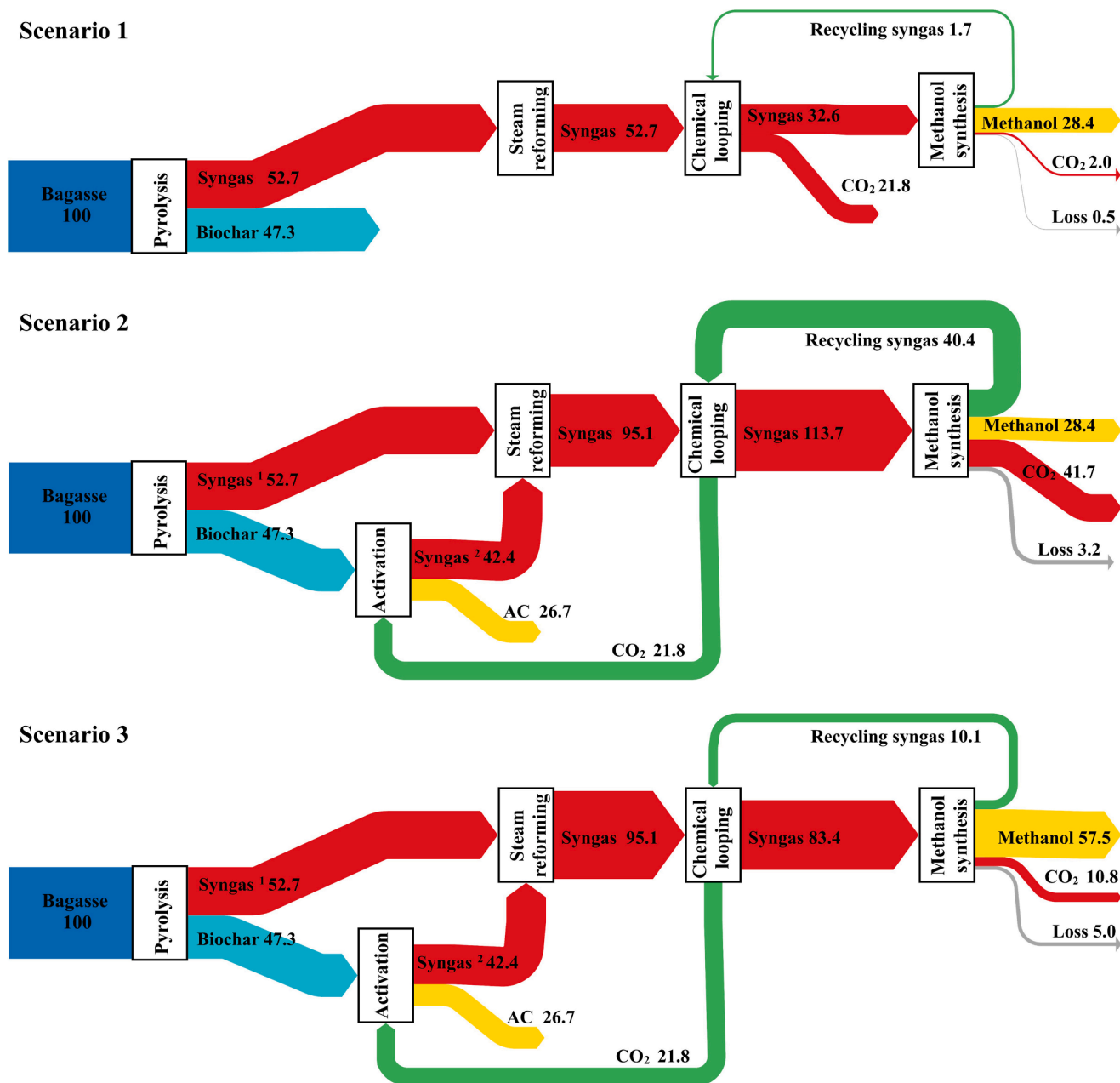


Fig. 4. Carbon flow of the three scenarios.

carbon production and quality, the amount of CaO was set as 5000 kg in the chemical looping process [23]. Fig. S4 presents the effect of input H₂ on the methanol yield in Scenario 3. The growth of input H₂ from 0 to 700 kg/h largely increased the methanol yield from 3703.11 to 9003.77 kg/h, as it could effectively improve the conversion ratio of carbon. Therefore, the amount of input H₂ was set to 500 kg/h in Scenario 3 from a conservative point. Moreover, Fig. S5 shows the syngas composition in each stage in the three scenarios. The H₂ content experienced a significant increase after the steam reforming process.

3.2. Carbon and energy efficiencies

Fig. 4 illustrates the carbon flow for the three scenarios. During the pyrolysis stage, bagasse was converted into conventional components, then 47.3% of the carbon remained in the biochar, with the rest in the form of syngas. The biochar was then converted into AC with CO₂ from the chemical looping process as an activator. Syngas generated from the

activation process was mixed with syngas from bagasse pyrolysis. The two syngas streams were subjected to the chemical looping process to adjust the gas composition. The improved syngas was then fed to the methanol synthesis process and converted to methanol. In Scenario 1, about 28.4% of the carbon was converted into methanol, and 47.3% of the carbon was retained in biochar. In Scenario 2, only 26.7% of the carbon was stored in AC due to the activation process. However, about 57.5% of the carbon was converted into methanol in Scenario 3 due to the addition of extra hydrogen. Table S4 lists the energy efficiency of the three scenarios. Those scenarios all achieved high energy efficiencies, and Scenarios 1 and 3 had a higher energy efficiency because of the high yield of biochar and methanol.

3.3. Techno-economic analysis

The economic feasibility and technological performance of the project are assessed through the TEA method. This evaluation can guide

Table 4
Techno-economic results of the three scenarios.

Items	Unit	Scenario 1	Scenario 2	Scenario 3
Input-fixed costs				
EC	KUSD ^a	4452.70	4956.30	5269.60
Pyrolysis and steam reforming unit	KUSD	1425.50	1425.50	1425.50
Activation unit	KUSD	–	503.60	503.60
Chemical looping unit	KUSD	1178.30	1178.30	1178.30
Methanol synthesis unit	KUSD	1848.90	1848.90	2162.20
IC	KUSD	7982.90	8924.80	9219.80
Pyrolysis and steam reforming unit	KUSD	2563.70	2563.70	2563.70
Activation unit	KUSD	–	941.90	941.90
Chemical looping unit	KUSD	2397.80	2397.80	2397.80
Methanol synthesis unit	KUSD	3021.40	3021.40	3316.40
PPC	KUSD	12435.60	13881.10	14489.40
TPC	KUSD	16166.28	18045.43	18836.22
TCC	KUSD	17782.91	19849.97	20719.84
WCI	KUSD	3556.58	3969.99	4143.97
OMC	KUSD	711.32	794.00	828.79
Input-variable costs				
Feedstock cost	USD/t ^b	37.79	37.79	37.79
Catalyst cost	USD/t	0.0407	0.0407	0.0407
CaO cost	USD	464.30	464.30	464.30
Transportation fee	USD/t	6.85	6.85	6.85
Hydrogen cost	USD/t	–	–	242.00
Input-unity costs				
Electricity cost	USD/t	16.85	16.85	19.01
Natural gas	USD/t	–	8.70	8.70
Water	USD/t	0.70	0.70	0.70
Output				
Methanol	USD/t	149.19	149.07	302.16
Biochar/AC	USD/t	96.16	158.18	158.18
Techno-economic indicators				
NPV	Million USD	80.43	110.49	47.78
IRR	%	6.53	5.80	9.13
PBP	Year	22.76	27.46	13.07

^a KUSD = 1000 USD.

^b USD/t: The cost or income of processing 1 ton bagasse.

project development, inform relevant research, assist capital investment decisions, and provide a basis for policy formulation. Table 4 lists the techno-economic results of the three scenarios. The TCC of Scenario 1 is the lowest because of the lack of activation equipment. In contrast, the TCC of Scenario 3 is the highest among the three scenarios, which is because the addition of hydrogen increases the methanol yield and the requirements of the methanol synthesis equipment. Meanwhile, the methanol synthesis unit is the largest contributor to capital investment among the three scenarios due to the high requirements for multi-tubular RPlug reactors of R11 and R12. The highest contribution of variable costs in Scenario 3 is hydrogen cost because of its high price. The unity cost with the greatest impact is electricity. The present pricing of methanol and AC leads to a low PBP of 6.53 years and a high IRR of 22.76% for Scenario 1 and a low PBP of 5.80 years and a high IRR of 27.46% for Scenario 2. However, the PBP of Scenario 3 is higher than the other two scenarios due to the high TCC and hydrogen price.

Sensitivity analysis is a vital tool to assess the robustness of the project and determine the impact of complicated relationships of various parameters on different scenarios. The parametric sensitivity analysis was performed to analyze the impact of variations in relevant factors on the decision-making objectives [24]. This study evaluated the effect of seven significant variables on the PBP. As presented in Fig. 5, the variation in hydrogen price had a tremendous impact on the PBP of Scenario 3, and the decline in hydrogen price decreased the PBP from 9.13 to 5.46 years. The Chinese government has set ambitious targets for reducing the cost of green hydrogen production, aiming to reach approximately 4000 USD/t by 2025 and 2400 USD/t by 2030 [59].

Predictably, the achievement of this goal will make Scenario 3 more feasible and attractive than Scenarios 1 and 2 under current conditions. The change in methanol price also largely affected the economic performance of Scenario 3 due to its high methanol yield. The variation in biochar/AC had a certain effect on their PBPs. Moreover, the fluctuation in capital cost exhibited a certain impact on those scenarios, but the risk of such a change is completely controllable. However, the variation in electricity, feedstock, and transport distance had a limited impact on their economic performance. Scenarios 1 and 2 exhibited greater robustness and resilience to risks from the changeable methanol and biochar or AC market due to its relatively low product yield and TCC. Given the current conditions and volatile markets, Scenarios 1 and 2 appear to be more favorable options, which can be selected according to market demand. However, the adjustment of the hydrogen input amount in Scenario 3 can flexibly regulate methanol yield to improve its economic benefits. Moreover, the reduction of green hydrogen production costs and the increment of electrolysis capacity in China will enhance the economic feasibility and competitiveness of Scenario 3. Therefore, Scenario 3 will replace the other two scenarios as the best option in the foreseeable future.

3.4. Life-cycle assessment

The meaningfulness of bioenergy production from biomass hinges on the attainment of a low carbon footprint under economically favorable circumstances [60]. Table 5 lists the LCA results of the three scenarios. The global warming potential (GWP) of the plantation was about 44.64 kg CO₂-eq/t, and each transport emitted 8.5 kg CO₂-eq/t. Moreover, the bagasse to methanol process consumed a lot of electricity and natural gas and released 311.49, 325.87, and 356.37 kg CO₂-eq/t for Scenarios 1, 2, and 3, respectively. However, methanol and biochar or AC could store most of the carbon from bagasse and lead to low GWPs of –993.38 and –542.12 kg CO₂-eq/t for Scenarios 1 and 2. Meanwhile, the hydrogen source significantly affected the environmental performance of Scenario 3, as displayed in Fig. 6 (a). Traditional hydrogen production technologies, such as SMR, S-I cycle, and EOW, entail significant resource and energy consumption, resulting in high GWP and greatly weakening the environmental performance of Scenario 3 [61]. However, the introduction of novel techniques and renewable energy and resources can largely mitigate CO₂ emissions or even further improve the environmental benefits of Scenario 3. As shown in Fig. 6 (b), Traditional methanol production technologies, including CTM and NTM, have high GWPs due to the huge consumption of fossil fuels. The introduction of green technologies, such as CCS and green hydrogen, can improve their environmental performance to a certain extent. However, BTM is a novel and revolutionary route to produce methanol with certain environmental benefits. Scenarios 1 and 2 achieved low GWPs of –1617.22 and –699.14 kg CO₂-eq/t methanol, which provided significant environmental benefits, especially in terms of reducing greenhouse gas emissions. Overall, each scenario has its own advantages and disadvantages, and it is necessary to make trade-offs and choices according to specific conditions. For instance, Scenario 1 could be prioritized if the main consideration is environmental protection. In contrast, Scenario 2 would be the first choice if the primary concern is economic growth. Similarly, if methanol yield and carbon efficiency are critical factors, then Scenario 3 might be favored.

3.5. Prospects and challenges

Sugarcane planting area in China has continued to increase in recent years, from 1.35 million hectares in 2015 to 1.48 million hectares in 2020 [68]. As a result, the annual sugarcane yield is approximately 70 million tons, generating about 20 million tons of bagasse [6]. According to the above results, bagasse resources can be converted into 7.41 million tons of methanol and 4.81 million tons of biochar in Scenario 1, 7.41 million tons of methanol and 2.78 million tons of AC in Scenario 2,

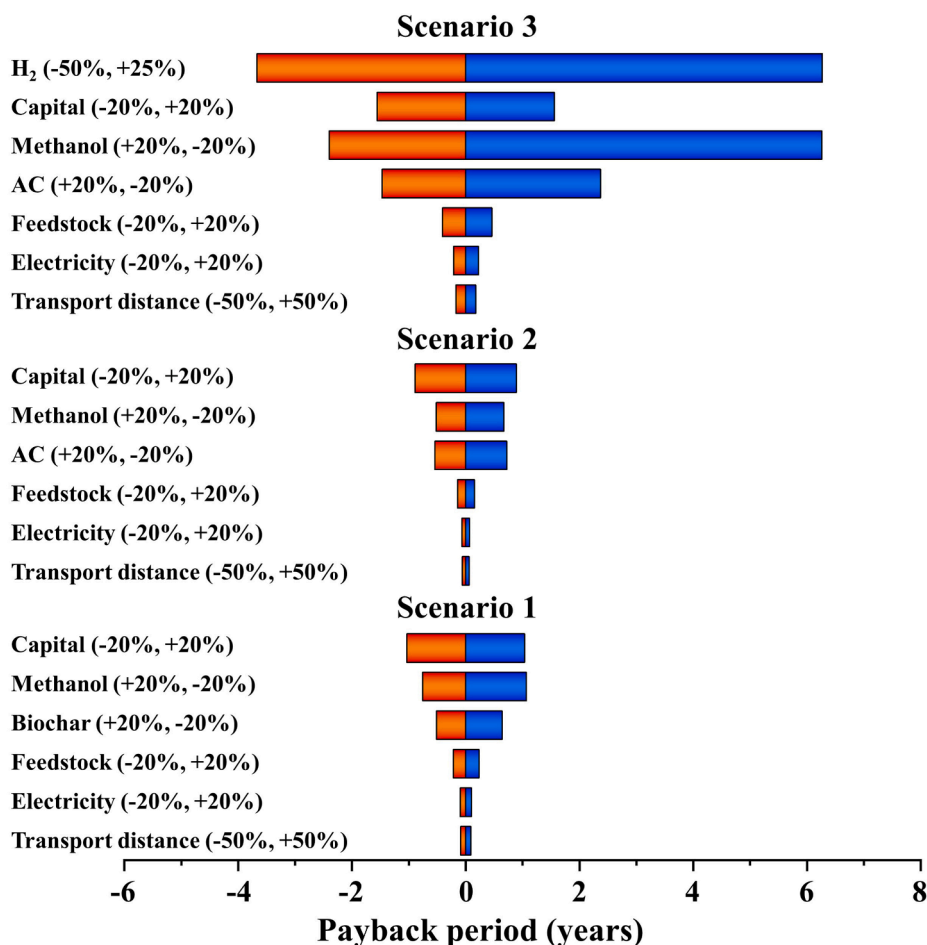


Fig. 5. Sensitivity analysis of seven variables to PBP.

Table 5
LCA results of the three scenarios.

Items	Unit	Scenario 1	Scenario 2	Scenario 3
Plantation				
Fertilizer	kg CO ₂ -eq/t	0.97	0.97	0.97
Water	kg CO ₂ -eq/t	37.91	37.91	37.91
Electricity	kg CO ₂ -eq/t	3.62	3.62	3.62
Diesel	kg CO ₂ -eq/t	2.14	2.14	2.14
Hydrogen	kg CO ₂ -eq/t	-	-	-4374.00–8280.00
Project				
Electricity	kg CO ₂ -eq/t	192.10	206.48	206.48
Natural gas	kg CO ₂ -eq/t	-	65.97	65.97
Transportation	kg CO ₂ -eq/t	8.50	8.50	8.50
Methanol	kg CO ₂ -eq/t	-509.18	-509.18	-1031.75
Biochar/AC	kg CO ₂ -eq/t	-848.83	-477.92	-477.92
GWP^a	kg CO ₂ -eq/t	-993.38	-542.12	3965.81 (SMR) 3545.81 (S-I cycle) 265.81 (Biomass)
GWP/methanol^b	kg CO ₂ -eq/t methanol	-1631.18	-710.28	232.52 (Biomass)

^a CO₂ equivalent reduction from the utilization of 1 ton of bagasse.

^b CO₂ equivalent reduction from the production of 1 ton of methanol, which is based on the method of economic allocation [62].

and 15.01 million tons of methanol and 2.78 million tons of AC in Scenario 3 theoretically. The total methanol production in China was 78.13 million tons in 2021, and this indicates that the implementation of Scenario 3 could contribute about 19.20% of the Chinese methanol yield annually. Given the current methanol and biochar/AC price, this project

can achieve considerable economic benefits of 4.91 billion USD in Scenario 1, 6.15 billion USD in Scenario 2, and 9.21 billion USD in Scenario 3. Based on the LCA results, the implementation of the project can mitigate 19.70 million tons of CO₂ in Scenario 1 and 10.67 million tons of CO₂ in Scenario 2. In 2021, the gross domestic product (GDP) of China amounted to 17734.06 billion USD, which means that the implementation of Scenario 3 could contribute roughly 0.052% of the Chinese GDP annually [69]. As for the environment, 11.47 billion tons of CO₂ were emitted in China in 2021; achieving successful implementation of Scenario 1 is projected to result in a reduction of approximately 0.17% of Chinese total annual CO₂ emissions, which is conducive to the realization of carbon neutrality [70].

The implementation and development of the project cannot be separated from the support of national policies. Numerous policies, regulations, and laws are issued and enacted to advance this process, such as the National Plan for Climate Change (NPCC) and the 14th Five-Year Plan (2021–2025). The NPCC sets targets for reducing CO₂ intensity by 40–45% from 2005 levels by 2020 [71]. China’s 14th Five-Year Plan includes several measures aimed at reducing CO₂ emissions and achieving carbon neutrality by 2060. To achieve this goal, the plan sets targets for decreasing the intensity of carbon emissions per unit of GDP by 18% compared to 2020 levels, raising the proportion of non-fossil fuels in primary energy consumption to 20%, and increasing forest coverage by 100 million hectares. The plan also prioritizes the development of renewable energy sources, including wind, solar, biomass, and hydropower, as well as the promotion of green transportation and the optimization of industrial structures. Additionally, the plan includes measures to promote carbon trading and carbon capture, utilization, and storage technologies [72]. This project is within the

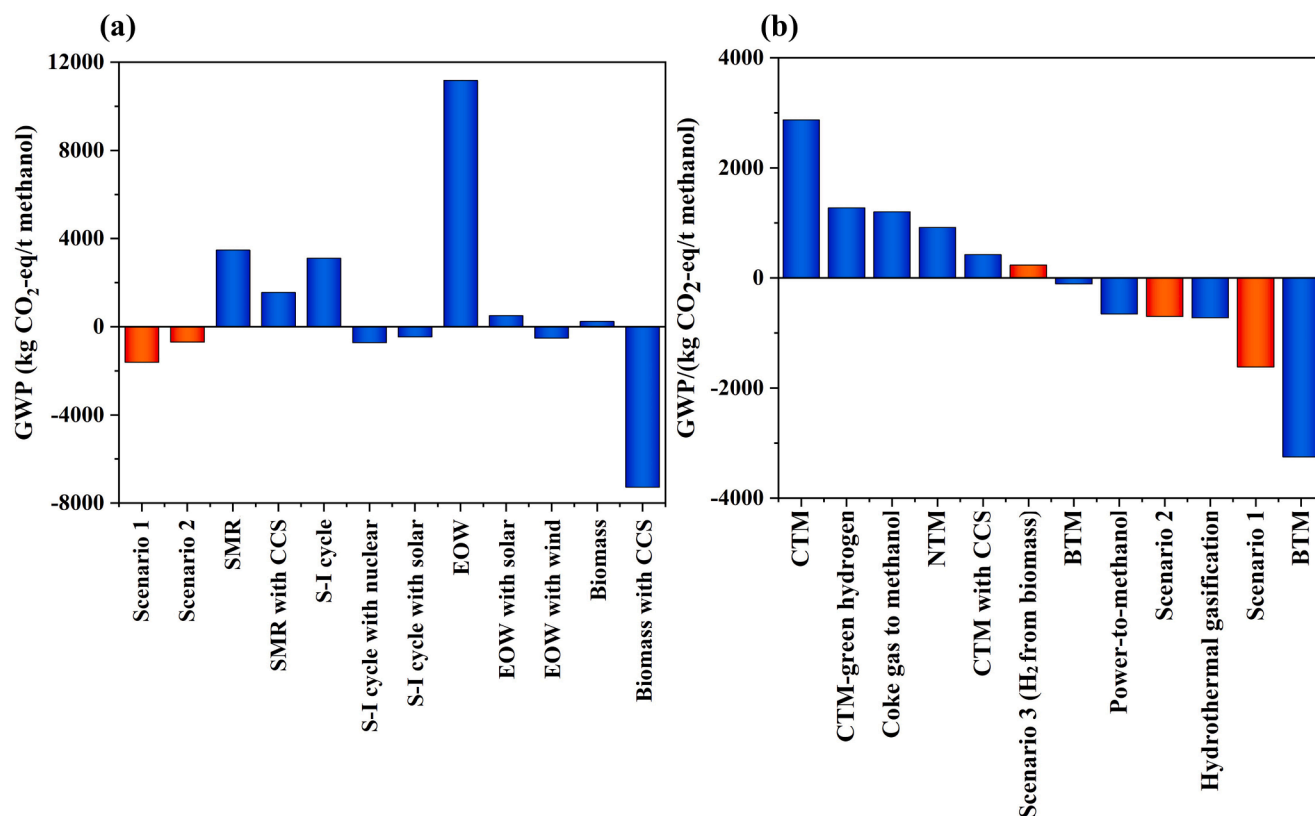


Fig. 6. Comparison of GWP results in (a) different hydrogen preparation methods and (b) different methanol production methods [38,63–67].

scope of policy support and will attract the attention of policymakers, shareholders, and researchers.

Additionally, the imposition of the carbon tax would further enhance the economic benefits of biomethanol. Presently, the carbon tax has been implemented by many counties, which is a policy that aims to make the social costs associated with carbon emissions visible. It incentivizes the use of cleaner and more sustainable energy sources by creating a more level playing field [73]. China does not impose a nationwide carbon tax system in place. However, there have been discussions and proposals to implement a carbon tax in China as a more comprehensive policy to address climate change. The government has conducted studies and pilot programs to evaluate the feasibility and potential impact of a carbon tax. It is foreseeable that the carbon tax will be fully implemented in China in the near future, which could provide a market-based incentive to reduce greenhouse gas emissions by making fossil fuels more expensive and incentivizing the use of cleaner and more sustainable energy sources [74]. By then, the production of biomethanol can be promoted as it has a lower carbon footprint compared to fossil-based methanol. The carbon tax makes fossil-based methanol more expensive, making biomethanol more economically competitive.

Furthermore, the utilization of CO₂ generated from the project is crucial for achieving sustainable and environmentally friendly industrial practices. The captured CO₂ can be utilized for various applications, such as microalgae cultivation, which has shown great potential in mitigating CO₂ emissions while producing valuable biomass. For instance, Banerjee et al. [75] demonstrated that microalgae (*Chlamydomonas reinhardtii*) could sequester 113 mg/L of CO₂ per day with high biomass productivity of 513 mg/L. For instance, utilizing the emitted 3921 kg/h of CO₂ with high purity from Scenario 1 for microalgae cultivation can lead to the daily production of 427.22 tons of microalgae. This approach not only helps to reduce CO₂ emissions but also has the potential to provide a sustainable source of biomass for various applications, such as animal feed, biofuels, and bioplastics.

Several challenges must be addressed to ensure its successful

implementation on an industrial scale. Given the widespread distribution of sugarcane plantations and sugar refineries across China, careful consideration is needed for the deployment and site selection of bio-refinery plants. Industrial processes must be integrated into the existing supply chain and logistics networks, with appropriate transportation, storage, and distribution arrangements in place [76]. This can require significant investment in logistics infrastructure, such as warehouses, shipping and distribution networks, and transportation systems. Transport distance and ecological protection are significant factors that impact transportation costs, project operation costs, and GHG emissions. Moreover, the scale of the methanol synthesis plant is also a crucial issue affecting the development of the methanol industry. While large-scale or centralized plants may have stronger bargaining power in the supply and sales chains, they still face many challenges, including high capital and operation costs, complex logistics, and high transportation fees. Conversely, small-scale or distributed plants have a lower economic threshold and more flexible transportation routes and logistics [77].

4. Conclusions

The study proposed a novel BTM method and investigated its economic and environmental benefits in China. Scenario 3 exhibited the highest carbon and energy efficiencies due to its high methanol yield, but Scenarios 1 and 2 are favorable options with low PBPs of 6.53 and 5.80 years. However, the decrease in green hydrogen production cost in China will enhance the economic feasibility and competitiveness of Scenario 3. Compared with traditional technologies, Scenarios 1 and 2 presented remarkable environmental benefits with low GWPs of −1631.18 and −710.28 kg CO₂-eq/t methanol. However, the environmental performance of Scenario 3 was heavily dependent on hydrogen sources, and the green hydrogen production method would significantly improve its environmental performance. Overall, it is necessary to weigh and choose among the three scenarios according to multiple factors, such as production goals, economic benefits, and environmental

impacts, to achieve the optimal production process. This study provides an economically viable and carbon-negative method for transforming low-grade biomass into eco-friendly methanol and AC, which is urgently required in the current background of achieving carbon neutrality.

CRediT authorship contribution statement

Guangcan Su: Conceptualization, Methodology, Investigation, Data curation, Formal analysis, Visualization, Validation, Writing – original draft. **Nurin Wahidah Mohd Zulkifli:** Supervision, Methodology, Validation, Data curation, Formal analysis, Funding acquisition, Project administration. **Li Liu:** Methodology, Validation, Resources, Data curation, Project administration, Software, Writing – review & editing. **Hwai Chyuan Ong:** Conceptualization, Supervision, Methodology, Resources, Writing – review & editing. **Shaliza Ibrahim:** Methodology, Resources, Supervision. **Yifan Wei:** Conceptualization, Validation, Data curation, Investigation. **Feng Bin:** Supervision, Resources, Writing – review & editing.

Declaration of Competing Interest

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

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.enconman.2023.117481>.

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