



Environmental and economic benefits of biomass-to-hydrogen systems through integrated catalytic steam reforming of pyrolysis bio-oil and co-product valorization

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ABSTRACT

Transitioning to a hydrogen-based economy requires renewable production pathways that can overcome environmental and economic limitations and become comparable to fossil-derived hydrogen. This study hypothesizes that process integration combined with co-product valorization can simultaneously improve environmental performance and reduce hydrogen production costs. Two biomass-to-hydrogen pathways are comparatively evaluated: a standalone catalytic steam reforming of pyrolysis bio-oil pathway (CRB) and an integrated pathway that couples bio-oil reforming and methane pyrolysis with biochar valorization (CRBB), using detailed process simulation, cradle-to-gate life cycle assessment, and techno-economic analysis. System CRBB achieves less environmental burdens in 16 out of 18 midpoint impact categories, 1.5 times lower cumulative energy demand than System CRB, and provides climate credits of -2.43 kg of carbon dioxide equivalent per kilogram of hydrogen through carbon black substitution. Economically, hydrogen production cost decreases from 6.18 to 2.86 dollars per kilogram, reaching as low as 1.12 dollars per kilogram under high-value biochar scenarios. These findings indicate that biochar valorization enables biomass-derived hydrogen systems to approach environmental and economic performance levels comparable to fossil-based hydrogen.

1. Introduction

Fossil fuels continue to dominate global primary energy consumption, but their long-term sustainability is increasingly challenged by resource depletion and environmental impacts. Today, more than 80% of global energy demand is met by coal, oil, and natural gas [1]. The societal reliance on these fuels, while historically essential for economic development, has led to severe challenges including greenhouse gas emissions, air pollution, and climate change [2]. As countries pursue decarbonization, green hydrogen has emerged as a promising alternative energy carrier because it emits no greenhouse gases when generated using renewable resources [3]. Hydrogen is gaining traction in transportation applications such as vehicles, trains, ships, and aviation, and it is also considered a viable substitute for natural gas in boilers, furnaces, and district heating systems for residential, commercial, and industrial buildings [4]. In this transition, biomass is expected to become an increasingly important and sustainable source for producing green hydrogen due to its renewability, carbon-neutrality, and wide availability [5].

Among biomass-to-hydrogen routes, catalytic steam reforming of fast-pyrolysis bio-oil has gained considerable attention because it couples a thermochemical conversion platform for green hydrogen production. Fast pyrolysis is a well-established and scalable approach for converting lignocellulosic biomass into valuable intermediates including bio-oil, char and non-condensable gases. In addition, it is widely regarded as a promising, sustainable, and relatively simple process compared to other thermochemical options [6]. However, despite extensive progress in pyrolysis technology, direct use of raw bio-oil as a fuel substitute remains challenging due to its high oxygen content, elevated viscosity, thermal instability, and corrosiveness [7]. Bio-oil typically contains up to 7 wt% hydrogen that can be extracted through steam reforming in the presence of metallic catalysts at elevated temperatures [8]. Upon condensation, pyrolysis vapor naturally separates into an organic phase rich in aromatic and lignin-derived compounds, and an aqueous phase rich in carbohydrate-derived oxygenates [9]. The former is devoted to the production of fine chemicals due to its high content of phenolic compounds, and the latter is the feed for catalytic steam reforming for hydrogen production due to its lower

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aromatic content and reduced coking tendency [10].

Catalytic steam reforming of bio-oil has been extensively explored at the unit-operation level, and numerous catalyst innovations have enhanced hydrogen yield and conversion efficiency. For example, Ce-incorporated Ni/Mg/Al catalysts have been shown to improve overall carbon conversion and raise hydrogen yield from 0.111 to 0.123 g H₂ per gram of organics in the feed, while simultaneously improving catalyst stability [11]. Precious-metal catalysts such as 5 wt% Pt on carbon black have demonstrated high H₂ selectivity of 87% when reforming representative aqueous-phase compounds such as levoglucosan, acetic acid, hydroxyacetone, furfural, formic acid, and methanol [9]. Other studies have shown that whole liquid fractions can be effectively converted with supplemental steam in the presence of heterogeneous reforming catalysts [8]. Ca-modified Ni/CeO₂•Al₂O₃ bifunctional catalysts have been developed to enhance the water–gas shift (WGS) reaction via CO₂ adsorption, thereby boosting hydrogen selectivity [12]. In parallel, extensive work has optimized pyrolysis conditions, product distributions [13], and reforming strategies [14].

While catalytic steam reforming of pyrolytic bio-oil has advanced significantly at the unit-operation level, combined life cycle assessment (LCA) and techno-economic analysis (TEA) of this pathway remains limited. Arfan et al. [15] evaluated hydrogen production from biomass and biowaste and reported that the environmental impact and life cycle cost of producing 1 kg of hydrogen were comparable. Their analysis identified steam reforming of pyrolysis bio-oil as a major environmental hotspot, accounting for more than 40% of impacts across seven categories, primarily due to electricity consumption. Other system-level investigations have focused predominantly on economic analysis. For example, Zhang et al. [16] demonstrated that hydrogen production via bio-oil reforming was economically more attractive than gasification, with a fixed capital investment approximately 88 million United States dollars lower and a broader internal rate of return range (8.9%–26.2%) compared to gasification (–2.6%–15.7%). Additionally, the majority of life cycle and techno-economic studies on biomass-to-hydrogen production have concentrated on gasification-based pathways. For instance, integration of carbon dioxide capture into gasification systems reduced environmental impacts but increased hydrogen minimum selling price by 0.1–0.3 dollars per kilogram depending on reactor configuration [17]. Similarly, Wu et al. [18] reported that the gasifier and air separation unit dominate capital investment, highlighting the importance of integrated system design rather than isolated component optimization.

Collectively, these studies provide valuable insights into process selection and design. However, they largely evaluate primary hydrogen generation pathways without systematically considering the environmental and economic contributions of co-products. In particular, biochar generated during biomass pyrolysis has typically been combusted for heat rather than valorized as a high-value product in prior analyses [15]. Earlier work treating biochar as a sole product showed that soil application can result in negative greenhouse gas emissions of –1.22 tonnes of carbon dioxide equivalent per tonne of dry biomass feedstock [19], demonstrating significant carbon abatement potential. More recently, biochar has been shown to possess both catalytic activity and high electrical conductivity. Wen et al. [20] demonstrated that microwave-assisted methane pyrolysis over renewable biochar enhances hydrogen production while producing carbon materials with exceptional electromagnetic shielding performance, achieving minimum reflection loss values as low as –73.1 dB. These findings suggest that biochar can play a multifunctional role in biomass conversion systems, serving both as a catalyst and as a precursor for advanced carbon materials.

Despite this potential, the environmental and economic implications of integrating biochar-catalyzed methane pyrolysis with bio-oil reforming have not been systematically evaluated within a unified modeling framework. Therefore, this study proposes a strategy that simultaneously enhances environmental performance and economic

viability of biomass-to-hydrogen production through process integration and co-product valorization. Two systems are comparatively assessed: a baseline pathway consisting of fast pyrolysis followed by catalytic steam reforming of pyrolytic bio-oil (CRB), and an integrated pathway that combines bio-oil reforming and methane pyrolysis with subsequent biochar valorization (CRBB). Using hemp hurds as a representative biomass feedstock, detailed Aspen Plus process simulations are developed and evaluated through cradle-to-gate life cycle assessment and techno-economic analysis. By linking environmental impacts and economic performance, this work quantifies the avoided burdens and cost offsets enabled by biochar valorization and provides system-level insights into how integrated biomass conversion pathways can deliver renewable hydrogen with improved sustainability and enhanced economic competitiveness.

2. Method

This study integrates process simulation, LCA, and TEA to evaluate two biomass-to-hydrogen pathways: (i) CRB and (ii) CRBB. Aspen Plus V14 was used to construct the model that provided consistent inputs to both LCA and TEA. The Aspen Plus property method was the Peng–Robinson equation of state with the Boston–Mathias modification (PR-BM) for thermophysical calculations. TEA estimates the minimum selling price (MSP) under discounted cash flow assumptions, while LCA followed ISO 14040 [21] /14044 guidelines [22] and was performed using SimaPro 10.2.0.3 coupled with the Ecoinvent 3.11 database. Three environmental indicators were quantified: ReCiPe 2016 Midpoint (H), cumulative energy demand (CED), and IPCC 2021 GWP100.

2.1. Process design and simulation

Two biomass-to-hydrogen pathways are modeled in Aspen Plus: (i) CRB: catalytic steam reforming of the aqueous fraction of fast-pyrolysis bio-oil; and (ii) CRBB: the same configuration integrated with methane pyrolysis, where pyrolytic biochar serves as a catalyst for CH₄ decomposition and the spent carbon is subsequently valorized (Fig. 1). In biomass pyrolysis simulation, hemp hurds were selected as the representative biomass feedstock due to their favorable availability. As a low-value byproduct of fiber hemp processing [23], hurds are produced in large quantities and do not compete directly with food or feed applications. According to the National Hemp Report by the U.S. Department of Agriculture, the United States has become a significant fiber hemp–producing country, with fiber hemp harvested from 5138 ha (12,690 acres) and yielding approximately 15 million kilograms (33.2 million pounds) of fiber hemp [24]. In both pathways, hemp hurds are subjected to fast pyrolysis, after which vapors are separated into biochar, noncondensable pyrolytic gas, aqueous bio-oil, and an organic fraction. The pyrolytic gas is combusted for heat supply. The aqueous fraction is directed to steam reforming to produce hydrogen, while the organic fraction is considered as a co-product. Hydrogen is then purified by pressure swing adsorption (PSA) and compressed for storage. The difference between CRB and CRBB lies in the treatment of pyrolytic biochar: in CRB, it is combusted to supply process heat, whereas in CRBB it is diverted to catalyze methane pyrolysis, generating additional hydrogen and the spent catalyst is a co-product that substitutes carbon black.

2.1.1. Pyrolysis

Fast pyrolysis was represented using a validated kinetic scheme for cellulose/hemicellulose/lignin decomposition [25], augmented with extractives reactions [26]. Model fidelity was checked against literature yields at 570 °C, showing acceptable agreement for gas, oil, and char (SI Table 1). Hemp hurds are the feedstock (composition from [27]); with ~10 wt% moisture reported and hurds are modeled as fully dried prior to conversion. The fractions including cellulose, hemicellulose, lignin, and extractives are calculated by the Solver function in Excel for the

supplied at 32,204.76 kg h⁻¹, producing CO₂ and H₂O to meet pyrolyzer and reformer duties. In CRBB, the biochar from hurd pyrolysis was diverted to methane pyrolysis, eliminating its combustion role and the solid carbon produced from methane pyrolysis was burned off in the combustor. The heat recycled from high-temperature gas streams was supplied to generate power. The summary of simulation results on heat duty in pyrolyzers, reformer, combustor, condenser and coolers is provided in SI Table 3.

2.1.7. Operating assumptions

Key assumptions for all major process units, including hurd pyrolyzer, condenser, reformer, combustor, methane pyrolyzer, and PSA, are summarized in Table 1.

2.2. Life cycle assessment

The life cycle assessment framework was developed to quantify the environmental impacts of hydrogen production through the two biomass-to-hydrogen pathways. This section describes the goal and scope definition, allocation procedures, inventory development, and impact assessment methodology.

2.2.1. Goal and scope

The goal of the LCA was to evaluate the environmental performance of hydrogen production from hemp hurds through the CRB and CRBB pathways under a cradle-to-gate framework. The functional unit was defined as 1 kg of hydrogen at 99.99% purity under standard temperature and pressure (STP). System boundaries included hemp cultivation, transportation, fast pyrolysis, bio-oil reforming, methane pyrolysis (for System CRBB), hydrogen purification, compression, and power generation (Fig. 1).

2.2.2. Allocation procedures

Mass and economic allocation. Since nearly all parts of hemp have potential uses [23], allocation of cultivation burdens among co-products

Table 1

Key operating assumptions for modeling the two hydrogen production pathways: CRB and CRBB. These values form the basis of the Aspen Plus simulations and subsequent LCA and TEA analyses.

Modeling Process	Parameters	CRB	CRBB	Ref.
Pyrolyzer 1	Hurd (dry weight), t/h	10	10	[37]
	Temperature, °C	600	600	Modeled
	Pressure, bar	1	1	Modeled
	Residence time, s	3.1	3.1	Modeled
	Biomass/N ₂	2:1	2:1	Modeled
	Biomass conversion rate	90%	90%	Modeled
	Condenser	Temperature, °C	50	50
Pressure, bar		6	6	Modeled
Reformer	Temperature, °C	650	650	[11]
	Pressure, bar	1	1	[11]
	Catalyst-to-oil ratio	0.057	0.057	[11]
	Modeled Products	H ₂ , CO ₂	H ₂ , CO ₂	Modeled
Combustor	Temperature, °C	500	500	[38]
	Pressure, bar	8	8	[38]
	air, kg/h	32204.76	32204.76	Modeled
	Modeled Products	H ₂ O, CO ₂	H ₂ O, CO ₂	Modeled
Pyrolyzer 2	Methane (Natural gas), kg/h		1500	Modeled
	Temperature, °C		600	[20]
	Pressure, bar		1	[20]
	CH ₄ /N ₂		3:2	[20]
	CH ₄ conversion rate		100%	Modeled
PSA 1	Absorption capacity of Zeolite 13X	2.62 mol CO ₂ /kg	2.62 mol CO ₂ /kg	[35]
	PSA 2	Absorption capacity of Zeolite 13X		5.22 mol N ₂ /kg

was required. Two approaches were applied: mass-based allocation and economic allocation. The allocation procedure follows the work [39]. For mass allocation, hemp biomass comprises 5% seed, 30% leaf, 20% fiber, and 45% hurds [40], yielding allocation factors of $\lambda_s = 0.05$, $\lambda_l = 0.30$, $\lambda_f = 0.20$, and $\lambda_h = 0.45$, where λ_s , λ_l , λ_f , λ_h , were the allocation factors for seed, leaf, fiber, and hurd, respectively.

For economic allocation, market values were assigned to seed (\$2.10/lb), fiber (\$1.99/lb), and hurd (\$0.45/lb), with no value attributed to leaves [39]. This resulted in allocation factors of 0.463 for seed, 0.438 for fiber, and 0.099 for hurd. When normalized to the total cultivation output of 15 t, the final allocation shares were $\lambda_s = 0.10$, $\lambda_f = 0.49$, and $\lambda_h = 0.29$. These factors were applied in sensitivity analysis to evaluate the effect of allocation choice on LCA results.

Avoided allocation. In System CRBB, pyrolytic biochar was utilized as a co-product (i.e., high-value electromagnetic shielding materials) after being used as a catalyst in methane pyrolysis. The avoided-burden method was applied, whereby the environmental credits from producing carbon black were subtracted from system impacts. The substitution is reasonable as the specific surface area of biochar can achieve 20.96 m²/g [41], comparable to carbon black (21.07 m²/g) [42]. Biochar derived from lignocellulosic biomass has been reported to exhibit high electrical conductivity and surface characteristics similar to those of commonly used conductive carbon black benchmarks [43], making it suitable for applications such as electromagnetic shielding and conductive composites. In addition, comparative studies have shown that biochar exhibits only a 17.5% lower reinforcing index than carbon black when used as a filler in styrene-butadiene rubber, indicating comparable mechanical reinforcement performance [44]. These material property similarities support the assumption of functional equivalence for substitution in this study. The influence of this assumption on life cycle results is further examined through sensitivity analysis in Section 3.4.

2.2.3. Life cycle inventory

The LCI was built using a combination of Aspen Plus simulation outputs and background datasets. The process chain was divided into six main stages: (i) hemp cultivation, (ii) hurd transportation, (iii) hurd conversion into H₂, (iv) methane pyrolysis (System CRBB only), (v) hydrogen purification and storage, and (vi) power generation. A summary of life cycle inventory inputs per functional unit is provided in Table 2.

2.2.4. Impact assessment methodology

The environmental impacts of the System CRB and System CRBB pathways were assessed using three indicators including ReCiPe 2016 Midpoint (H), cumulative energy demand (CED), and IPCC 2021 GWP100. Detailed explanation about these indicators see SI Table 4. All results were expressed relative to the functional unit of 1 kg of hydrogen at 99.99% purity under STP. Biogenic CO₂ uptake during hemp growth was credited to the system, while subsequent release during processing was reported separately from fossil CO₂ emissions. In the CRBB system, substitution credits were applied using the avoided-burden approach, where spent biochar displaced carbon black.

2.3. Techno-economic analysis

The techno-economic analysis was conducted to evaluate the economic viability of the CRB and CRBB pathways under consistent financial and operational assumptions. A discounted cash flow rate of return (DCFROR) framework was employed to estimate the MSP of hydrogen. The MSP was defined as the selling price at which the net present value (NPV) of the project equals zero over the assumed plant lifetime. This approach ensures that the calculated hydrogen price reflects the long-term financial sustainability of each pathway.

The analysis assumed a 30-year plant lifetime with a 10% IRR, consistent with capital-intensive energy projects. Additional financial

Table 2

Life cycle inventory inputs for the functional unit (1 kg of hydrogen at 99.99% purity, STP) for the two modeled pathways: CRB and CRBB. Inventory data were obtained from Aspen Plus simulation outputs, literature sources, and Ecoinvent/technical reports as specified. Negative values indicate avoided burdens credited through co-product substitution (e.g., biochar replacing carbon black in System CRBB).

Process units	Unit	Inputs for System CRB	Inputs for System CRBB	Data sources
Hemp cultivation				
Hemp	kg	39.11	25.26	Simulation result
Hurd transportation				
Distance	mile	500	500	Map
Hurd	kg	16.00	10.34	Simulation result
Hurd pyrolysis and bio-oil reforming				
Hurd	kg	16.00	10.34	Simulation result
CO ₂ biogenic	kg	13.62	5.03	Simulation result
Bio-oil	kg	8.15	5.26	Simulation result
Reforming catalyst (loading)	kg	0.002	0.001	Simulation result [11]
Water	kg	3.55	2.29	Simulation result
Electricity (natural gas)	kWh	5.96	3.85	Simulation result
CO ₂ biogenic	kg	10.66	6.89	Simulation result
Carbon black	kg	0	-1.03	Simulation result
Methane pyrolysis				
Methane	kg	0	1.56	Simulation result
Electricity (natural gas)	kWh	0	3.13	simulation result
CO ₂ fossil	kg	0	3.87	simulation result
H ₂ purification and storage				
Electricity (natural gas)	kWh	0.53	0.53	[45,46]
Zeolite	Kg	0.003	0.002	simulation result
Power generation				
Electricity (natural gas)	kWh	0.58	0.40	[46]

parameters included an income tax rate of 21%, 100% equity financing, and 8000 annual operating hours. The construction period was modeled as 2.5 years, with expenditures distributed as 8% in the first 6 months, 60% in the following year, and 32% in the final year. Start-up was assumed to last 6 months, during which revenues and variable costs were set to 75% of nominal values, and fixed costs were maintained at 100%. Working capital was taken as 5% of total capital investment, and land acquisition costs were assumed to equal 6% of purchased equipment costs. These parameters refer to [46] and are summarized in Table 3.

Capital expenditures (CAPEX) were estimated based on purchased equipment costs obtained from literature and technical reports. The costs of equipments on pyrolysis, reforming and power plant set-up refer to [46]. The cost of feedstock pretreatment refers to [37]. The expense of cooling utilities refers to [38]. Costs were scaled to the plant design capacity:

$$C_{\text{scale-up}} = C_{\text{base}} \times \left(\frac{\text{Scale-up capacity}}{\text{Base capacity}} \right)^k$$

where C_{base} is the reference cost, capacity scaling was based on process throughput, and k is the scaling exponent. All costs were corrected to 2020 U.S. dollars using the Chemical Engineering Plant Cost Index (CEPCI):

Table 3

Key economic parameters applied in the techno-economic analysis of hydrogen production from hemp hurds. These values were used in the discounted cash flow rate of return (DCFROR) model to estimate the minimum selling price of hydrogen for both CRB and CRBB pathways.

Economic parameters	Value
Internal rate of return	10%
Income tax rate	21%
Equity	100%
Plant life	30 years
Annual operating hours	8000 h
General plant depreciation	7 years
Construction period	2.5 years
spent in first 6 months expenditures	8%
spent in next 12 months expenditures	60%
spent in last 12 months expenditures	32%
Start-up time	6 months
Revenue:	75%
Variable costs:	75%
Fixed costs:	100%
Working capital	5% of total capital investment
Land	6% total purchased equipment cost

$$C_{\text{corrected}} = C_{\text{scale-up}} \times \frac{\text{CEPCI}_{2020}}{\text{CEPCI}_{\text{base}}}$$

The total capital investment (TCI) was then calculated as the sum of scaled and corrected equipment costs, indirect costs, working capital, and land.

Operating expenditures (OPEX) were divided into variable and fixed costs. Variable costs included feedstock (hemp hurds and natural gas), electricity, catalysts, and waste treatment. Fixed costs accounted for labor, maintenance, insurance, and overhead. Revenues from co-products were explicitly included as credits: in CRB, the organic bio-oil fraction was modeled as a potential fine-chemical feedstock, while in CRBB, both the organic bio-oil fraction and spent biochar are co-products at market prices. The catalyst cost is estimated using CatCost model [37] assuming that catalyst replacement frequency is ten times cycle of deactivation-activation. Metal (Earth Abundant) on Metal Oxide is selected as the template process in alignment with the selected raw materials and catalyst preparation method in the paper [11].

Finally, heat integration was conducted to minimize external energy demand. The resulting reduction in net heating and cooling requirements improved overall system efficiency as well as heat recovery for power generation, decreasing extra electricity demand. Details of the integration results are summarized in SI Table 3. The analysis assumes that the generated power is enough to supply auxiliary electricity for feed pretreatment, pumping, and PSA operation except H₂ compression and electricity for operating the power generation unit because H₂ compression accounts for the largest power use [46]. The proportion of heat loss and conversion loss of electricity to thermal energy accounted for 42% of the actual input electricity [47].

2.4. Sensitivity and scenario analysis

Sensitivity and scenario analyses were performed to evaluate the robustness of the environmental and economic results and to identify the parameters with the greatest influence on system performance. Both LCA and TEA outcomes were examined under a series of alternative assumptions to capture uncertainties in feedstock supply, energy inputs, natural gas supply, co-product markets, and process parameters. Selection of sensitivity analysis parameters can be seen in SI Table 5.

3. Results and discussion

This section presents and interprets the environmental and economic performance of the two biomass-to-hydrogen pathways. The discussion

begins with life cycle environmental impacts, followed by cumulative energy demand, global warming potential, sensitivity analysis, and techno-economic performance.

3.1. Life cycle environmental impacts

The comparative life cycle assessment of the two biomass-to-hydrogen systems (CRB and CRBB) reveals clear differences in their environmental performance across the 18 midpoint impact categories considered under the ReCiPe 2016 framework. Fig. 2 summarizes the relative contributions of each system, where negative values indicate avoided burdens from co-product or environmental benefits and positive values reflect environmental impacts. Overall, System CRBB demonstrates a lower environmental burden than System CRB in 16 out of 18 categories, except GWP and HNCT. These improvements are attributed to the integration of methane pyrolysis and the subsequent valorization of biochar, which provide both additional hydrogen output and credits for avoided production of carbon black.

Across 11 categories including SOD, OFH, PMF, OFTE, TA, FE, ME, TET, FET, LU, WC, even without the avoided burden of carbon black substitution, the impact of System CRBB is lower than System CRB. This

is due to the process combination of methane pyrolysis and hurd pyrolysis, weakening the impact of the component ‘hurd conversion’ that finally accounts for over 60% of the impact distribution. Hydrogen production relies entirely on biomass conversion, meaning that all cultivation, transportation, and pyrolysis burdens are allocated to the production of one kilogram of hydrogen. In the integrated system, a portion of hydrogen is generated from methane pyrolysis. As a result, the biomass throughput required per kilogram of hydrogen decreases, effectively diluting the environmental burden associated with hurd conversion. On the other hand, thanks to avoided burdens the net impact of System CRBB is markedly decreased to below 100% of System CRB in IR, ME, HCT, MRS, FRS. The impact intensity for HCT, with avoided burden, was reduced from 245.22% to 89.97%, resulting in a lower impact than System CRB and reducing carcinogenic toxicity to humans. The environmental credits of co-product contribute to compensating environmental burden regarding IR, ME, HCT, MRS, FRS, imposed by the integration of methane pyrolysis, mainly caused by natural gas production and methane pyrolysis. For HNCT, both systems make positive benefits to the environment.

Among all impact categories, System CRBB only exhibits slightly higher global warming potential (GWP100) compared to System CRB,

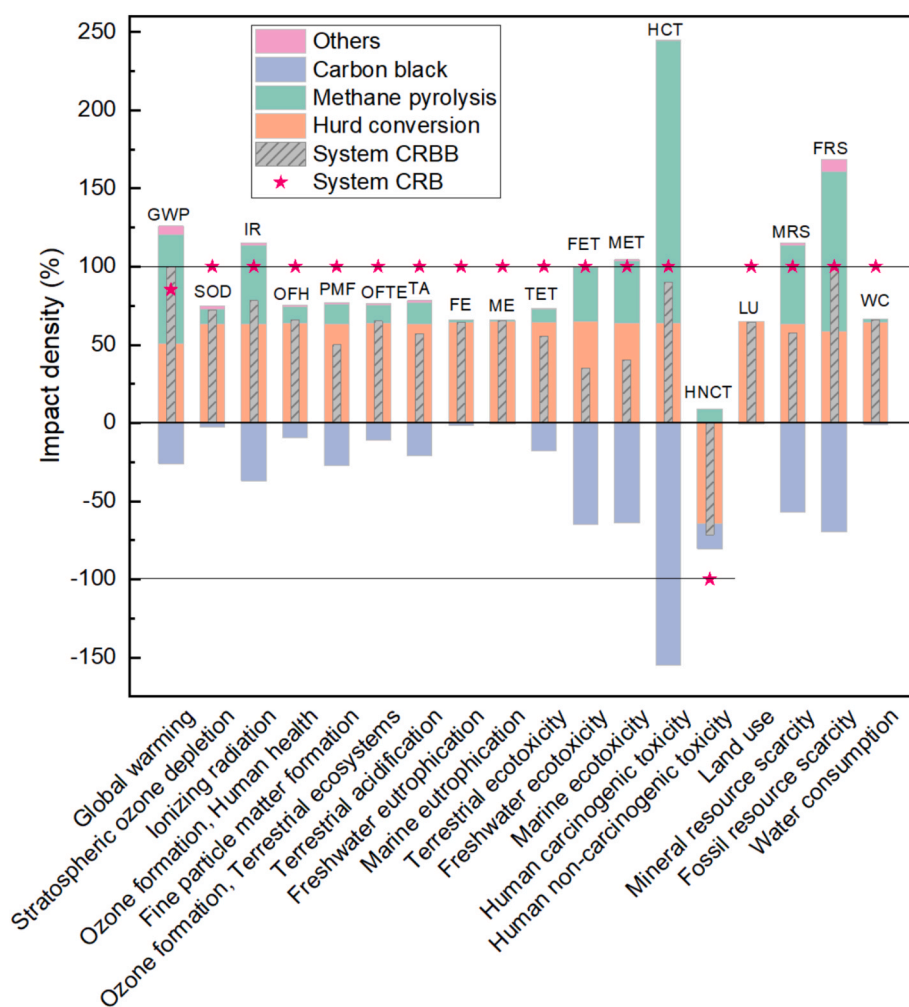


Fig. 2. Comparison of environmental impacts across 18 ReCiPe 2016 Midpoint (H) categories for System CRB and System CRBB. Results are normalized to the functional unit (1 kg H₂ at 99.99% purity) and expressed as percentage contributions relative to System CRB. In the bar chart, positive values represent environmental burdens, while negative values indicate avoided burdens or environmental credits. The “hurd conversion” category in System CRBB includes hurd cultivation, hurd transportation, hurd pyrolysis, and bio-oil reforming. In the figure, star markers (★) indicate the impact values for System CRB, and hatched bars represent the net impacts of System CRBB. The colored segments decompose contributions by process stage in System CRBB: hurd conversion (orange), methane pyrolysis (blue-green), carbon black credit (blue), and the other category representing electricity for H₂ storage and power generation (purple). Overall, System CRBB reduces impacts in 16 out of 18 categories, except GWP and HNCT, although GWP is slightly higher compared to System CRB.

primarily due to fossil CO₂ emissions from methane pyrolysis. This slight difference is partially offset by the avoided burden of biochar substitution, which provides a climate credit of up to 2.43 kg CO₂-eq/kg H₂, lowering the net emissions of System CRBB close to those of System CRB. The analysis result is similar to that of the study that showed gasification systems can achieve net-negative greenhouse gas (GHG) emissions when electricity co-products are credited against fossil power [17]. The impact from the ‘others’ category can be neglected, as it occupies only a small percentage in System CRBB.

Taken together, these results suggest that the integration of methane pyrolysis with biochar valorization can transform the environmental performance of biomass-to-hydrogen systems. System CRBB achieves broader environmental benefits beyond climate impacts, although its competitiveness in GWP100 is contingent on the extent of biochar substitution credits. In contrast, System CRB lacks this offsetting mechanism, making its environmental profile more uniformly burdensome across categories.

To validate generalizability of the conclusions, comparison of environmental impacts across 18 ReCiPe 2016 Midpoint (H) categories for System CRB and System CRBB using wheat straw as biomass feedstock was indicated in SI Fig. 1. The figure indicates that System CRBB exhibits a lower environmental burden than System CRB in 17 out of 18 impact categories. Owing to the lower hydrogen yield and greater biochar production with more avoided burden, the GWP of System CRBB is 1.51 kg CO₂-eq per kg H₂ lower than that of System CRB, more clearly demonstrating the relative advantage of the integrated CRBB pathway. Overall, CRBB consistently outperforms CRB, reinforcing the robustness of the central conclusion that process integration and biochar valorization enhance environmental performance regardless of specific biomass composition.

3.2. Cumulative energy demand

The CED analysis provides additional insight into the energy

efficiency of the two hydrogen production pathways. As shown in Fig. 3, System CRBB demonstrates a markedly lower renewable energy requirement compared to System CRB. Specifically, the renewable energy demand of System CRB is 128.48 MJ/kg H₂, nearly 1.5 times higher than that of System CRBB, 83.37 MJ/kg H₂. This difference arises because System CRB relies exclusively on biomass as the primary feedstock for hydrogen production, whereas System CRBB offsets part of its renewable energy demand through methane pyrolysis.

In terms of nonrenewable energy inputs, System CRBB exhibits a higher baseline consumption due to its dependence on natural gas for methane pyrolysis and increased electricity use. However, when considering the associated co-product credits, the non-renewable energy requirement of System CRBB decreases significantly, from 254.80 MJ to 121.48 MJ/kg H₂, which is slightly lower than the value for CRB (123.50 MJ/kg H₂). This outcome highlights the critical role of co-product allocation in shaping the comparative energy performance of the two systems.

Overall, System CRBB achieves a more balanced energy profile by reducing total CED despite integrating fossil-based methane pyrolysis. The avoided burdens of biochar not only mitigate the added fossil energy use but also provide a net reduction in total energy demand compared to System CRB. This suggests that coupling biomass pyrolysis with biochar valorization can enhance energy efficiency while enabling greater flexibility in feedstock sourcing.

3.3. Global warming potential

Fig. 4 presents the breakdown of greenhouse gas emissions for System CRB and System CRBB expressed as GWP100. In System CRB, the largest contributor to CO₂ emissions is the hurd pyrolysis and bio-oil reforming stage, which accounts for more than 41.79% of total emissions. This is primarily due to electricity consumption associated with the production and use of natural gas. Hurd transportation is the second-largest contributor, reflecting the use of diesel fuel for long-distance

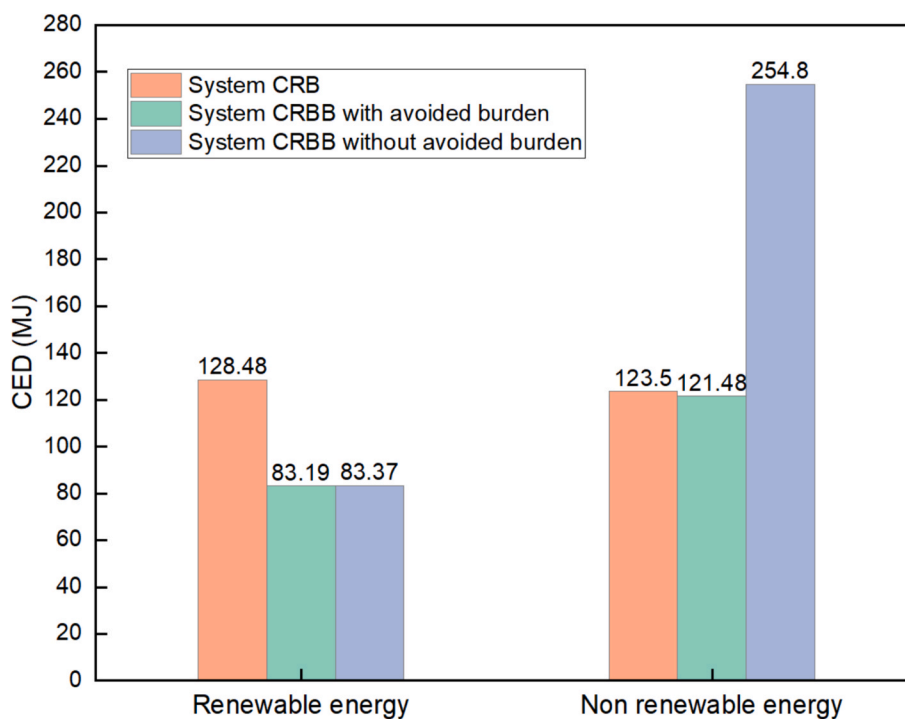


Fig. 3. Cumulative energy demand (CED) for CRB and CRBB. Results are shown for two categories: renewable and non-renewable energy demand. The legend distinguishes System CRB (orange), System CRBB with avoided-burden credit (green), and System CRBB without avoided-burden credit (blue). Values are normalized to 1 kg H₂ at 99.99% purity. CRBB achieves lower nonrenewable energy demand compared to System CRB, particularly when biochar substitution for carbon black is credited, while renewable energy use remains comparable across systems.

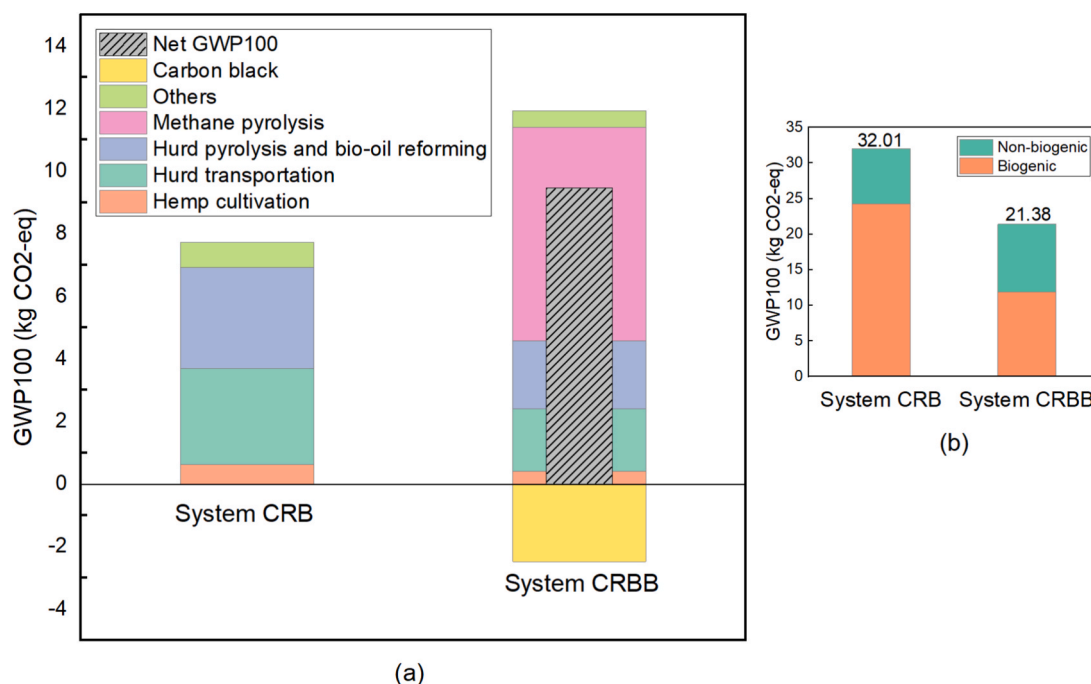


Fig. 4. (a) Global warming potential (GWP, 100-year time horizon, IPCC 2021) for hydrogen production from System CRB and System CRBB. Contributions are disaggregated into hemp cultivation, hurd transportation, hurd pyrolysis and bio-oil reforming, methane pyrolysis, carbon black substitution, and other minor sources (electricity for H₂ storage and power generation). Negative contributions indicate avoided burdens, such as carbon black substitution in System CRBB. System CRBB reduces net GWP100 relative to System CRB when co-product valorization is included, despite higher direct emissions from methane pyrolysis. Fig. 4. (b) The GWP 100 from biogenic and non-biogenic sources. The biogenic emissions are CO₂ produced from biomass conversion (see inventory).

biomass logistics. Together, these stages dominate the life-cycle GWP of System CRB, resulting in net emissions of 6.31 kg CO₂-eq/ kg H₂.

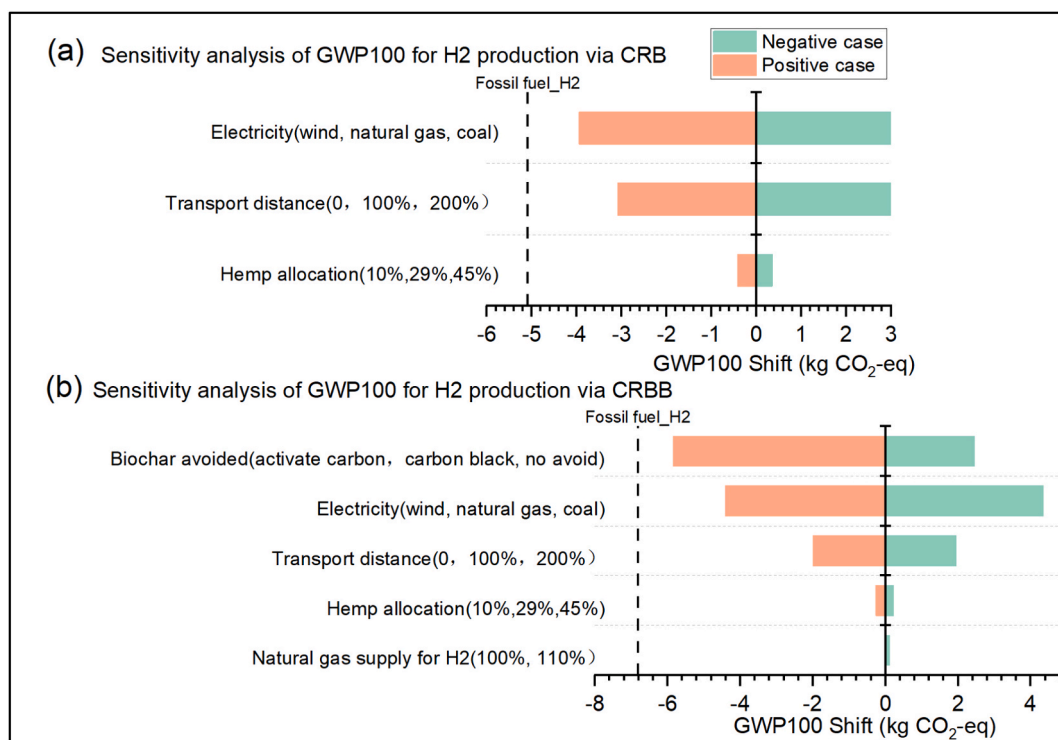


Fig. 5. Sensitivity analysis of GWP100 for H₂ production via System CRB and System CRBB. The tornado chart illustrates the effect of varying key parameters including feedstock transport distance, electricity source, allocation approach (mass- vs. economic-based), and biochar substitution scenarios, on net GWP100 outcomes. The middle value or parameter in parentheses is the base case. Positive cases (orange bars) indicate parameter changes that increase emissions relative to the baseline, while negative cases (green bars) indicate reductions. The CO₂ emission factor of fossil-fuel H₂ is 2.64 kg CO₂-eq/kg H₂ as reference. System CRBB results are particularly sensitive to assumptions on avoided allocation and electricity source, while System CRB impacts are more strongly driven by electricity source and feedstock logistics.

In System CRBB, the addition of methane pyrolysis substantially changes the emissions profile. Methane pyrolysis contributes 6.8 kg CO₂-eq/kg H₂, representing over half of the total gross emissions of the system. This increase is primarily attributable to fossil CO₂ emissions associated with natural gas production and combustion related to methane pyrolysis. Although the avoided burden from biochar substitution provides significant climate credits of − 2.43 kg CO₂-eq/kg H₂, these credits do not fully offset the added fossil emissions. Consequently, the net GWP100 of System CRBB (9.46 kg CO₂-eq/kg H₂) remains slightly higher than that of System CRB (7.73 kg CO₂-eq/kg H₂). However, regarding to total GWP 100, System CRBB exhibits 10.63 CO₂-eq/kg H₂ lower than System CRB as indicated in Fig. 4 (b). Biogenic CO₂ emissions are accounted for separately due to CO₂ absorption during hemp growth, so slightly higher GWP of System CRBB arises from added fossil emissions associated with methane pyrolysis rather than from biomass-derived carbon, despite the presence of avoided burdens.

Overall, the results emphasize that while System CRBB introduces additional fossil-related emissions, co-product valorization plays a decisive role in improving its climate performance. Importantly, the avoided-burden credit plays a decisive role in improving its climate performance, making the GWP100 of CRBB competitive with that of CRB, and effectively alleviating the greenhouse effect generated by methane pyrolysis.

3.4. Sensitivity analysis of GWP

To better understand the robustness of the life cycle results, a sensitivity and scenario analysis was performed on key parameters influencing the GWP100 of both System CRB and System CRBB, as shown in Fig. 5. For System CRB, the most influential factor is the source of electricity. Replacing natural gas with wind power for electricity reduces emissions by 3.94 kg CO₂-eq/kg H₂, highlighting the importance of decarbonizing process energy inputs. Transport distance is the second-most sensitive parameter, as longer transportation routes increase diesel consumption and associated emissions. In contrast, allocation methods for hemp co-products (mass- versus economic-based) have only a marginal effect on System CRB's overall GWP, indicating that the selection of allocation method in the cultivation stage has no significant effect on the LCA result.

For System CRBB, the most critical determinant of climate performance is the treatment of biochar substitution. When substituting activated carbon for carbon black, System CRBB achieves a GWP of 3.6 kg CO₂-eq/kg H₂, which is lower than base case (9.5 kg CO₂-eq/kg H₂) and closer to fossil-fuel H₂ (2.64 kg CO₂-eq/kg H₂). If no avoided burden is allocated to biochar, the GWP of System CRBB rises further to 11.9 kg CO₂-eq/kg H₂, exceeding System CRB. Electricity type also has some influence, but its effect is secondary compared to co-product assumptions. Feedstock allocation choices play a minor role. Even under this lower-price assumption (10% allocation), the resulting changes in allocation factors lead to only minor variations in GWP100, indicating that the life cycle environmental results are robust across a realistic range of regional and temporal hemp co-product price fluctuations. In addition, the GWP100 impact of extra 10% natural gas supply demonstrates negligible differences across scenarios indicating that the climate performance of the integrated system is not strongly sensitive to deviations from the assumed complete methane conversion.

Overall, these results underscore that System CRB's climate performance is primarily dependent on decarbonizing energy supply and reducing transport emissions, whereas System CRBB's performance hinges on the market value and substitution potential of biochar. This highlights both an opportunity and a risk: System CRBB can offer substantial climate benefits if biochar is successfully valorized, but its competitiveness diminishes without credible substitution pathways.

Notably, the potential ecological impacts associated with large-scale biochar production warrant careful consideration. Excessive removal of agricultural residues or biomass for pyrolysis may reduce soil organic

carbon inputs, alter nutrient cycling, and negatively affect long-term soil fertility if not managed properly [48]. Sustainable harvesting guidelines that retain a portion of residues in the field, combined with nutrient management strategies and site-specific biomass removal limits, are essential to mitigate these risks. Prioritizing low-value residues from forestry and agricultural systems, rather than dedicated energy crop overharvesting, can further reduce ecological pressure. While these considerations highlight important trade-offs, they do not negate the environmental benefits observed in the integrated pathway, but rather emphasize the need for responsible biomass sourcing within sustainable land management frameworks.

3.5. Techno-economic results

The techno-economic analysis reveals distinct cost structures between CRB and CRBB. Fig. 6 compares the total installed equipment costs for both systems. Both systems share several key units (hurd pretreatment, hurd pyrolysis, steam reforming, PSA, etc.), but CRBB also includes methane pyrolysis as an additional cost component. Due to the same hurds input, the installed equipment costs of hurd pretreatment, hurd pyrolysis, combustion, condenser, and steam reforming are the same. The power generation infrastructure dominates the capital cost structure both in CRB (39.51%) and CRBB (35.82%) as the sum cost of multiple equipment including extraction steam turbine, hot process water softener system, coolers, and pumps are relatively high along with their installation costs calculated with installation factor of 2.47 [46]. By comparison, other units mainly compose of reactors expense. The sensitivity of this unit to MSP is discussed in the following section. The cost difference between the two systems is attributed to integration of methane pyrolysis into SBRR. Although methane pyrolysis requires new reactor capacity and downstream purification, its contribution to total CAPEX remains modest at only 5.54% due to the relatively small scale of methane input. As a result, total capital investment for CRBB is estimated at \$144.50 million, compared to \$121.60 million for CRB, reflecting an 18.80% increase primarily from the methane pyrolysis unit and PSA equipment.

Annual operating costs show clear contrast between the two systems (Fig. 7). Both systems have the same expenses, such as feedstock, catalysts, and waste disposal due to the same hurd input in line with the capital cost analysis mentioned above. CRBB incurs higher variable costs because of additional natural gas requirements and greater electricity use for methane pyrolysis and H₂ compression. However, this increase is offset by the revenue from biochar credits. When biochar is valorized as a carbon black substitute at a market price of \$2.0/kg, CRBB achieves a net operating cost of \$18.34 M/year, nearly 36.54% lower than the \$28.90 M/year of CRB. This demonstrates that biochar revenue streams are the dominant differentiator in system-level economics, making CRBB more economical than CRB.

The resulting minimum selling price of hydrogen is summarized in Fig. 8. CRB produces hydrogen at an MSP of \$6.18/kg, which is substantially higher than the benchmark fossil-derived hydrogen cost of \$1.15/kg. This renders CRB economically unattractive without major cost reductions. By contrast, CRBB achieves a baseline MSP of \$2.86/kg, nearly three times lower than CRB, owing to its biochar co-product credits. Under a goal-case scenario in which biochar is valorized as a high-value co-product at \$3.70/kg, the MSP of CRBB decreases further to \$1.12/kg, comparable to fossil-based hydrogen.

Fig. 9 illustrates the sensitivity of MSP to key cost parameters. For CRB, feedstock cost dominates system economics, with H₂ MSP shifting between \$4.74/kg and \$7.65/kg. In contrast, CRBB is most sensitive to biochar value, which can reduce MSP to \$1.12/kg depending on market conditions and co-product quality. Notably, without revenue from biochar, the MSP in CRBB is \$4.91/kg, lower than that in CRB, which suggests that process integration delivers economic benefits. As indicated in this work [49], integrating pyrolysis intermediate streams with a traditional petroleum refinery decreases MSP by 32%. Other variables,

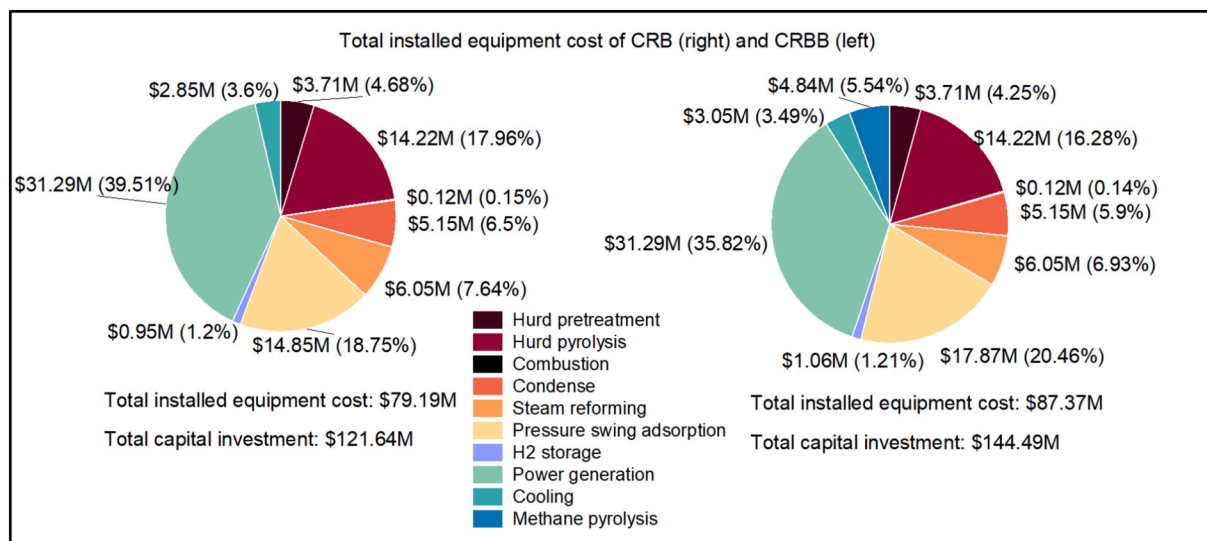


Fig. 6. Breakdown of total installed equipment cost for hydrogen production via CRB and CRBB. Costs are allocated across major process units, including hurd pretreatment, hurd pyrolysis, combustion, condensation, steam reforming, methane pyrolysis (CRBB only), Pressure swing adsorption, H₂ storage, power generation, and cooling. Colors correspond to equipment categories as shown in the legend.

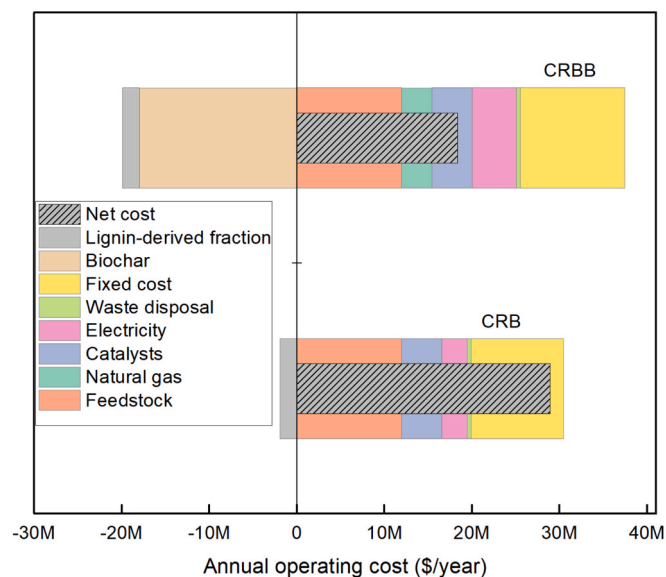


Fig. 7. Annual operating cost breakdown for CRB and CRBB. Costs are categorized into feedstock, natural gas, catalysts, electricity, waste disposal, fixed costs, and credits from co-products (biochar and lignin-derived fraction). Net cost is represented by the hatched bar. CRBB incurs higher operating expenses due to methane pyrolysis and natural gas input, but also benefits from biochar valorization credits, which reduce the overall net cost compared to CRB.

such as IRR, electricity price, and natural gas cost, exert secondary influences but do not fundamentally change system competitiveness. In CRBB, the variation of catalyst costs has the least effect on H₂ MSP.

Overall, the TEA highlights that CRB alone is neither cost-competitive nor resilient to feedstock price fluctuations, whereas CRBB leverages biochar valorization to achieve both lower MSP and greater economic flexibility. The decisive role of biochar revenue suggests that future research and policy support should prioritize developing high-value biochar markets, such as electromagnetic shielding materials, activated carbon, or carbon nanostructures, to ensure the long-term economic viability of integrated biomass-to-hydrogen pathways.

While the present work provides a system-level comparative

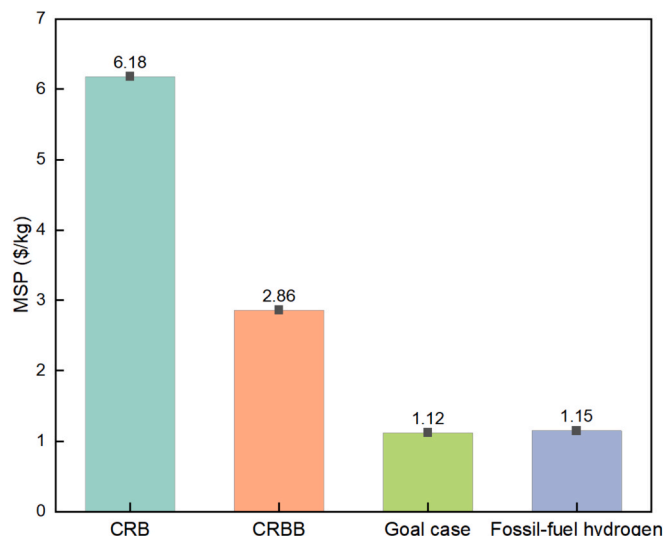


Fig. 8. Minimum selling price (MSP, \$/kg H₂) of hydrogen for different production pathways. Results are shown for CRB, CRBB, a “goal case” scenario reflecting potential process improvements, and fossil-fuel hydrogen as a benchmark. MSP values were determined using discounted cash flow analysis with a 10% internal rate of return over a 30-year plant lifetime. Integration of methane pyrolysis and biochar valorization in CRBB reduces the MSP relative to CRB, while the goal case approaches being competitive with fossil-based hydrogen.

assessment based on literature-validated models, future studies should further explore experimental validation of integrated operation and catalyst stability under continuous conditions. Additional process optimization and heat-integration refinement may further enhance system performance and economic competitiveness.

4. Conclusion

This study presented a comparative life cycle and techno-economic assessment of two biomass-to-hydrogen pathways based on hemp hurds: catalytic steam reforming of pyrolysis bio-oil and catalytic steam reforming with biochar valorization. The analysis demonstrated that

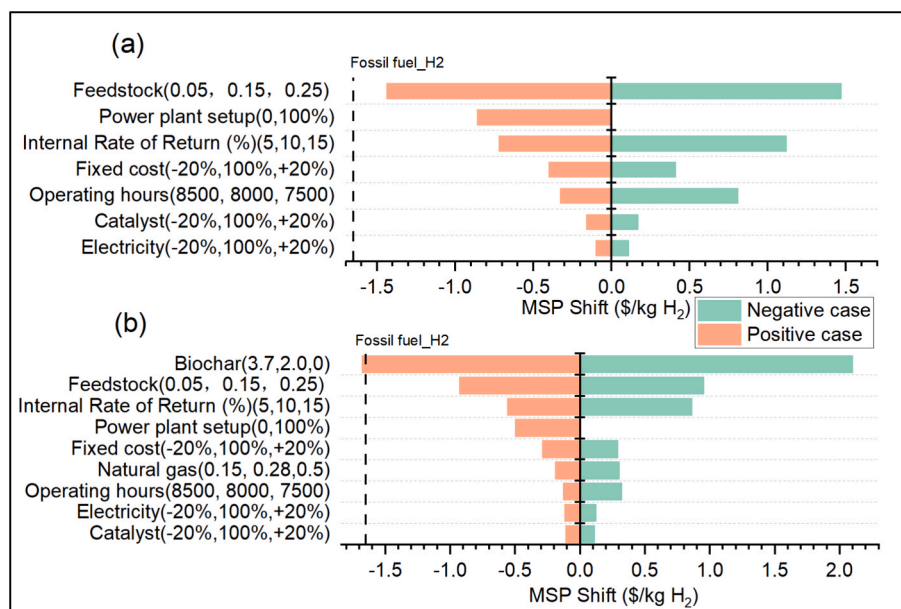


Fig. 9. Sensitivity analysis of hydrogen MSP (\$/kg H₂) for CRB and CRBB. The middle value in parentheses is the base case. Positive and negative cases represent parameter variations that increase or decrease the base MSP, respectively. Key parameters include feedstock cost, natural gas price, electricity price, catalyst cost, fixed operating cost, and co-product credits. MSP for CRB is most sensitive to feedstock, while CRBB is strongly influenced by the market value of biochar.

while CRB is limited by high environmental burdens and poor cost competitiveness, CRBB offers significant improvements through the integration of methane pyrolysis and the valorization of biochar as a co-product.

From an environmental perspective, System CRBB achieved a lower environmental burden than CRB in 16 out of 18 categories except GWP and HNCT. These improvements are attributed to the integration of methane pyrolysis and the subsequent valorization of biochar, which provide both additional hydrogen output and credits for avoided production of carbon black. Cumulative energy demand analysis demonstrates System CRBB reflects lower reliance on non-renewable fossil and consumed much less renewable energy. In terms of GWP100, the avoided burden of co-product provides significant climate credits of -2.43 kg CO₂-eq/kg H₂ that are associated with the production of carbon black. After accounting for these credits, the net emissions of CRBB decrease to 9.46 kg CO₂-eq/kg H₂, which is slightly higher than the System CRB value (7.73 kg CO₂-eq/kg H₂) but within a comparable range. Sensitivity analysis further highlighted the dependence of System CRBB's environmental benefits on the type and value of co-product substitution, underscoring the importance of credible market pathways for advanced biochar applications.

From an economic perspective, when biochar is valorized as a carbon black substitute at a market price of $\$2.0/\text{kg}$, CRBB achieves a net operating cost of $\$18.34$ M/year, nearly 36.54% lower than the $\$28.90$ M/year of CRB. CRB produced hydrogen at a minimum selling price of $\$6.18/\text{kg}$, well above the cost of fossil-derived hydrogen ($\$1.15/\text{kg}$), making it economically unviable. By contrast, CRBB achieved a baseline MSP of $\$2.86/\text{kg}$, and as low as $\$1.12/\text{kg}$ under a high-value biochar scenario, thereby approaching cost parity with fossil-based hydrogen. Sensitivity analysis revealed that CRB's economic is dominated by feedstock price, whereas the MSP in CRBB is most sensitive to biochar market value. However, even without revenue from biochar, the MSP in CRBB is $\$4.91/\text{kg}$, still lower than that in CRB, highlighting the economic advantage of process integration.

For broader context, it is important to position the present results relative to other low-carbon hydrogen pathways. Water electrolysis depending on renewable electricity has been reported to achieve GWP values typically 2.23 kg CO₂-eq/kg H₂ when supplied by wind and solar power, but would increase to 6.25 kg CO₂-eq/kg H₂ when grid electricity

is used [50]. Biomass gasification pathways have demonstrated when coupled with carbon capture, but often require higher capital investment and complex gas-cleaning infrastructure [17]. Compared to these alternatives, the integrated biomass-to-hydrogen pathway evaluated in this study offers competitive climate performance and economic potential when biochar valorization is realized, while leveraging solid carbon co-products that are not available in electrolysis-based systems.

In conclusion, the integration of biochar valorization into biomass-to-hydrogen systems presents a promising strategy to simultaneously improve environmental performance and economic feasibility. CRBB emerges as a potentially competitive renewable hydrogen pathway to replace fossil-fuel H₂, provided that biochar markets mature and technology improvements-such as microwave-assisted pyrolysis, can be scaled effectively. Future work should focus on optimizing biochar upgrading, expanding its application portfolio, and coupling biomass-based hydrogen production with renewable energy sources to maximize both climate benefits and economic resilience.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work, the authors used chat.openai.com to check the grammar. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

CRedit authorship contribution statement

Maoshui Zhuo: Writing – review & editing, Writing – original draft, Validation, Methodology, Formal analysis, Data curation, Conceptualization. **Sydney Byres:** Writing – review & editing, Data curation. **Yuanzhe Liang:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Investigation, Funding acquisition, Conceptualization.

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.

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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.2026.121314>.

Data availability

Data will be made available on request.

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