

1      **An integrated techno-economic and environmental assessment for carbon**  
2      **capture in hydrogen production by biomass gasification**

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7

8      **Abstract**

9      Bioenergy with carbon capture and storage (BECCS) is a potential solution addressing climate  
10     change, regional wildfires, and circular economy. This study investigates the economic and  
11     environmental performance of a BECCS pathway implementing carbon capture (CC) in hydrogen  
12     production via gasifying forest residues in the American West, by developing a framework that  
13     integrates process simulations, techno-economic analysis (TEA), and life cycle assessment (LCA).  
14     The results show that forest residue-derived hydrogen is economically competitive (\$1.52–2.92/kg  
15     H<sub>2</sub>) compared with fossil-based hydrogen. Incorporating CC increases environmental impact due  
16     to additional energy and chemical consumption, which can be mitigated by the energy self-  
17     sufficiency design that also reduces CC cost to \$75/tonne of CO<sub>2</sub> for a 2,000 dry short ton/day  
18     plant, or using renewable energy such as solar and wind. Compared to electrolysis and fossil-based  
19     routes with CC, only BECCS can provide carbon-negative hydrogen and is more favorable  
20     regarding human health impact and near-term economics.

21

22      **Keywords**

23      Carbon capture, Hydrogen, Biomass gasification, Techno-economic analysis, Life cycle  
24      assessment, BECCS

25

26     **1. Introduction**

27     Bioenergy with carbon capture and storage (BECCS) has the potential to limit global warming  
28     by providing net negative greenhouse gas (GHG) emissions (Donnison et al., 2020). The Sixth  
29     Assessment Report recently published by IPCC (Intergovernmental Panel on Climate Change)  
30     estimated the global cumulative CO<sub>2</sub> removal from BECCS from 2020 to 2100 to be as high as  
31     30–780 Gt CO<sub>2</sub> (IPCC, 2021), contributing to the Paris Agreement's target to limit the temperature  
32     increase to 1.5 °C (Torvanger, 2019). Furthermore, BECCS provides a non-fossil energy  
33     alternative and is vital in promoting energy security (Fajardy and Mac Dowell, 2018). The energy  
34     and climate benefits of BECCS have led to increasing interest in the research, development and  
35     deployment of BECCS, e.g., biomass carbon removal and storage (“BiCRS”) systems in the United  
36     States (Fajardy et al., 2019; Galik, 2020; New Energy and Industrial Technology Development  
37     Organization, 2021; Rosa et al., 2021).

38     BECCS cover various biochemical (e.g., ethanol fermentation) and thermochemical  
39     conversion pathways (e.g., combustion, gasification, and pyrolysis (Bui et al., 2021; Cheng et al.,  
40     2021; Hanssen et al., 2020)). Compared to biochemical pathways, thermochemical pathways have  
41     many advantages, such as lower purification requirements and higher flexibility in feedstocks,  
42     products, and scalability (Sanchez and Kammen, 2016; Shahbaz et al., 2021). Among different  
43     thermochemical pathways, gasification is promising in fuel decarbonization and supporting  
44     circular economy (Nunes, 2022). Gasification thermally breaks down biomass into syngas, a  
45     mixture of gases such as CO, CO<sub>2</sub>, and H<sub>2</sub>. H<sub>2</sub> is an essential industrial gas in the oil and chemical  
46     industry and a carbon-free fuel (Salkuyeh et al., 2018). Hydrogen from biorenewable sources is  
47     considered more environmentally preferable than hydrogen made from fossil fuel resources. It was  
48     estimated that solid biomass in the United States can supply 48 million metric ton (MMT) of  
49     hydrogen per year (Connelly et al., 2020), which are larger than the hydrogen demand estimated  
50     in the literature (22 to 41 MMT/year) (U.S. DOE, 2020a). Given the high tolerance of  
51     heterogeneous biomass feedstock, gasification has been explored to convert various waste  
52     materials (e.g., municipal solid wastes) to H<sub>2</sub> and valuable chemicals as a circular economy  
53     enabling technology (Bhatia, 2014). The process is also less prone to emission problems (e.g.,  
54     sulfur-containing emissions compared to the flue gas from post-combustion systems) for carbon  
55     capture (CC) since gas cleaning is already an essential part of the process (Neubauer and Liu,  
56     2013). Some regional assessments show the advantages of gasification. For example, Baker et al.  
57     (2020) assessed different negative emissions pathways (natural solution, BECCS, and direct air  
58     capture) and concluded that gasification of the solid biomass types to produce hydrogen has the  
59     largest promise for CO<sub>2</sub> removal at the lowest cost in California. Given the growing interest in the  
60     circular economy and the urgent need for decarbonization, gasification-based BECCS to produce  
61     hydrogen shows great promise in contributing to a more sustainable, circular, and low-carbon  
62     society, yet needs more understanding for its impacts directed to these potentials.

63     Previous studies have used techno-economic analysis (TEA) or life cycle assessment (LCA)  
64     to assess the economic feasibility and environmental impacts of gasification-based BECCS  
65     (Andrea Corti, 2005; Ghiat et al., 2021; Oreggioni et al., 2017; Rhodes and Keith, 2005; Valente  
66     et al., 2019). These studies have focused on biomass-based integrated gasification combined cycle

67 (IGCC) systems with post-combustion CC. A systematic literature review on TEA and LCA of  
68 biomass gasification for hydrogen production is provided in Supplementary Materials (SM)  
69 Section 1 Literature Review. The review shows that most TEA and LCA studies of gasification  
70 systems focus on hydrogen production without considering CC. Several studies have mentioned  
71 the importance of electricity sources in biomass co-gasification/gasification systems (Arnaiz del  
72 Pozo et al., 2021a; Martín-Gamboa et al., 2016), yet the effect of energy supply choices and  
73 strategies have not been fully explored. For example, Susmozas et al. (2016) show that adding CC  
74 contributes to negative carbon impact but at the price of worse life cycle environmental impacts  
75 compared to the system without CC. The authors highlighted the improvement opportunities of  
76 minimizing external electricity demand and direct air emissions, although the study does not  
77 analyze specific strategies. BECCS systems consume energy not only in the hydrogen production  
78 steps such as gasification, product cleaning, and hydrogen purification (Ahmed et al., 2012), but  
79 also in the carbon capture steps, which are typically energy-intensive (Roussanaly et al., 2021).  
80 The means to provide energy (e.g., internal generation or external supply, renewable or fossil) are  
81 important to determine the economics (Arnaiz del Pozo et al., 2021b) and environmental impacts  
82 (Susmozas et al., 2016) associated with energy consumption. In addition, few studies (Antonini et  
83 al., 2021) have considered impact categories beyond climate impact. Process-level analysis for  
84 different energy supply strategies and a broad range of impact indicators are essential in  
85 understanding the practical decarbonization role of BECCS with a consideration of other  
86 environmental impacts.

87 Not all CO<sub>2</sub> emission sources within a biorefinery are considered in previous LCA and TEA  
88 of BECCS systems. For example, Antonini et al. (2021) investigated the life cycle environmental  
89 impact of hydrogen production from wood gasification systems by taking the hydrogen end-use  
90 into account, which shows possible negative total GHG emissions for fuel cell electric vehicles  
91 using hydrogen from biomass. This study includes CO<sub>2</sub> from syngas but not CO<sub>2</sub> from gas cleaning  
92 off-gas and energy generation (i.e., steam and electricity). Salkuyeh et al. (2018) performed a TEA  
93 and an LCA to compare different gasification systems with CC. Their system captured CO<sub>2</sub> from  
94 syngas and flue gas stream of the steam and power generation but did not capture CO<sub>2</sub> in gas  
95 cleaning and gasification off-gas. Susmozas et al. (2016) conducted an LCA for hydrogen derived  
96 from short-rotation poplar biomass through gasification coupled with CC, and the study only  
97 includes CO<sub>2</sub> from the exhaust gas of the boiler. Both Salkuyeh et al. (2018) and Susmozas et al.  
98 (2016) captured CO<sub>2</sub> emissions of internal energy generation, but they have not explored how  
99 different energy supply strategies would affect CO<sub>2</sub> capture and overall decarbonization potential  
100 of their BECCS systems. Holistic understandings of the complex interactions between energy  
101 supply strategies and CC implementation are critical to simultaneously maximize the carbon  
102 removal potential and energy efficiency of BECCS.

103 This study addresses these knowledge gaps by developing an integrated techno-economic-  
104 environmental assessment (TEES) framework. The framework integrates process simulations,  
105 TEA, and LCA for gasification-based BECCS using forest residues prevalent in the Pacific  
106 Northwest U.S., where large volumes of biomass are available, and there is a pressing need to thin  
107 forests to mitigate severe wildfire. Sensitivity and scenario analyses were conducted to identify  
108 critical driving factors and understand the impacts of different energy supply strategies. The

109 analysis includes all CO<sub>2</sub> emission sources within a gasification biorefinery and explored how  
110 different energy supply strategies would affect CO<sub>2</sub> capture potential and costs. Process-level TEA  
111 and LCA examined how recent and future carbon prices and renewable energy access might  
112 incentivize BECCS deployment and affect the economic and environmental performances of  
113 different system design. In addition to climate impact, the LCA includes other environmental  
114 impact categories such as human health, eutrophication, acidification, ecotoxicity, and others.  
115 Although this study focuses on forest residues, the knowledge generated from this study can inform  
116 future research and large-scale deployment of BEECS for other waste feedstocks or in other  
117 regions.

## 118 2. Methods

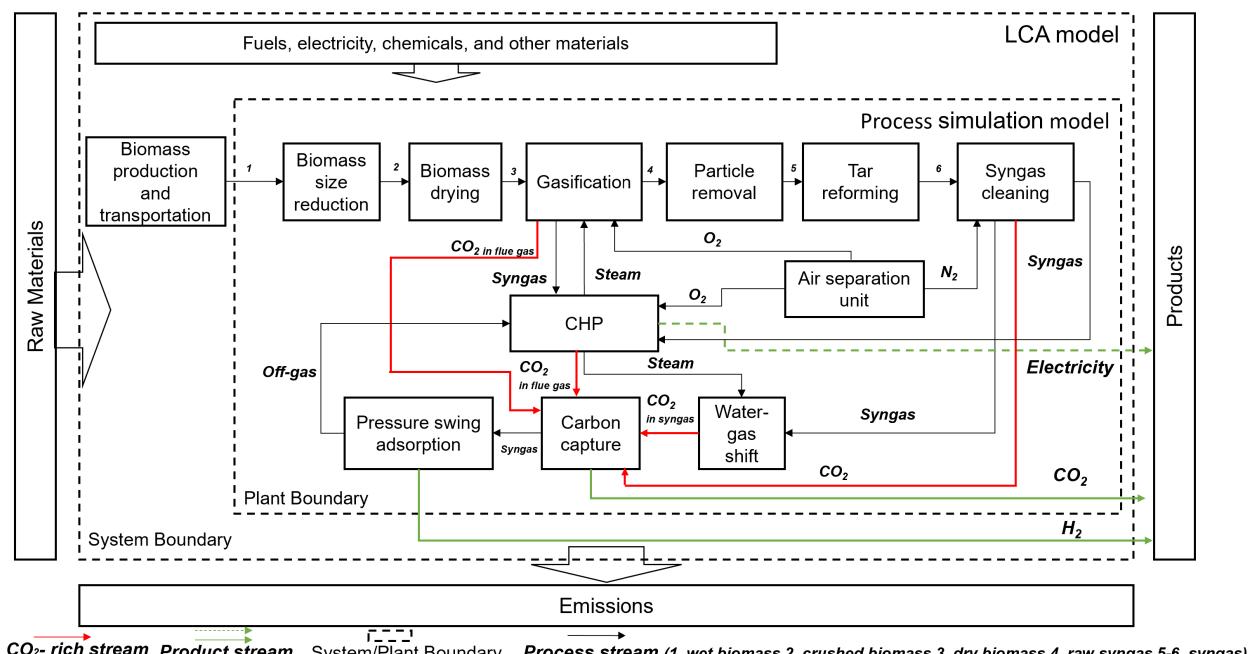
119 The TEES framework connects mass and energy flows from process simulation with  
120 engineering economics for TEA and life cycle inventories (LCI) for LCA. The process model was  
121 developed in Aspen Plus V11 (AspenTech, 2022). The detailed mass and energy balances from  
122 engineering rigorous process simulations provide physically sound data for TEA and LCA (Wu et  
123 al., 2021). A discounted cash flow rate of return analysis was conducted to calculate economic  
124 metrics, including CAPEX, OPEX, and minimum selling price (MSP) of H<sub>2</sub> and CO<sub>2</sub> (which is  
125 equivalent to the levelized cost of CC in this study (Lan et al., 2021a). A scenario analysis was  
126 developed to address the CC integration and energy supply strategies, which are often overlooked  
127 or insufficiently addressed in previous research (Roussanaly et al., 2021). For each scenario, TEA  
128 and LCA were performed to evaluate the process efficiency, financial performance, and life cycle  
129 environmental impacts. Moreover, a sensitivity analysis was applied to determine the effects of  
130 parameter variations on economic performances. The system boundary includes biomass  
131 production and transportation and all the unit operations in the biorefinery. CO<sub>2</sub> transportation and  
132 storage after CC is not considered.

### 133 2.1. Process description and simulation

134 Fig. 1 shows the system boundary of the BECCS system in this study. The process model  
135 within the plant boundary includes eight component subsystems: biomass preparation (size  
136 reduction and drying), gasification, syngas clean-up, water-gas shifting, CC, pressure swing  
137 adsorption, air separation unit, and heat power generation. After the feedstocks arrive at the plant,  
138 forest residues are crushed to reduce the particle size and dried from 25 wt% moisture (Cao et al.,  
139 2020; Haarlemmer, 2015; Motta et al., 2018) down to 10 wt% on a wet basis (Ståhl et al., 2004;  
140 Svoboda et al., 2009) to be suitable for gasification. After biomass preparation, dried forest  
141 residues are fed to the gasification system. The gasification system employs a dual fluidized bed  
142 gasifier/reactor (DFBR) consisting of a gasifier and a combustor. The combustor oxidizes the  
143 residual char from the gasifier to provide heat for the endothermic gasification reactions in this  
144 study. The gasifier and combustor are interconnected by circulating bedding-olivine with catalyst  
145 MgO (Spath and Ringer, 2005). Steam is used as the gasifying agent. The produced syngas and  
146 solids exit the gasifier and flow to the cyclone separators, where particulates are removed from the  
147 hot gas. The removed particles such as ash are landfilled. The raw syngas leaving the cyclone then  
148 passes through the tar removal system, where the tars and other unsaturated hydrocarbon  
149 compounds are converted into hydrogen and carbon monoxide using the alumina-based catalyst.

150 Syngas is then further cooled through a heat exchanger by cooling utilities. The Rectisol process®  
 151 (developed by Linde and Lurgi) (Burr and Lyddon, 1998; Kohl and Nielsen, 1997; Taheri et al.,  
 152 2018) is used to clean the syngas by removing acidic gas (sulfur) for syngas cleaning (see details  
 153 in Section 2.1.2). The excess scrubber water is sent for wastewater treatment. After syngas clean-  
 154 up, partial syngas (30%) is sent to combined heat and power generation (CHP) for power self-  
 155 sufficiency, depending on the scenarios that are discussed in Section 2.1.3. The rest of the syngas  
 156 goes through the water-gas shifting process, which includes the high-temperature shift (HTS) and  
 157 low-temperature shift (LTS) to convert CO and water into CO<sub>2</sub> and H<sub>2</sub>. The gas mixture then goes  
 158 to the CC section, which uses amine-based scrubbing solvents for CO<sub>2</sub> capture. To obtain high  
 159 purity hydrogen (99.9 vol%), a pressure swing adsorption (PSA) unit is used to separate the  
 160 impurities such as CO<sub>2</sub>, and CO, CH<sub>4</sub>, and other hydrocarbons. The off gas from PSA is sent to  
 161 CHP for energy recovery. For the combustion procedures (gasification and CHP), oxyfuel is  
 162 deployed using an air separation unit (ASU) to enrich the CO<sub>2</sub> concentration in the flue gas  
 163 (Borgert and Rubin, 2013; Kather and Kownatzki, 2011). The air separation unit also provides  
 164 nitrogen to the Rectisol process for CO<sub>2</sub> and H<sub>2</sub>S separation. The CO<sub>2</sub>-enriched flue gas from  
 165 gasification and CHP are also sent to the CC section to obtain purified CO<sub>2</sub> (more than 99.5 mol%).

166



167 **Fig. 1.** The system boundary of hydrogen production by biomass gasification with CC. The  
 168 system boundary represents the boundary of LCA and TEA. Plant boundary represents the  
 169 boundary of process simulation for the biorefinery.  
 170

171

### 172 2.1.1. Biomass characteristics

173 Forest residues in the Pacific Northwest region are used as biomass feedstock, specifically,  
 174 Douglas fir and Ponderosa pine (mass ratio 1:1 in this study). The average composition of forest  
 175 residues is shown in Table 1.

176

177 **Table 1.** The average composition of forest residues used in this study.

Components	Value
<b><i>Proximate analysis (wet basis, w%)</i></b>	
Moisture	25.00
Fixed carbon	13.05
Volatile matter	61.50
Ash	0.45
<b><i>Ultimate analysis (dry basis, w%)</i></b>	
Carbon	52.34
Hydrogen	6.35
Oxygen	40.54
Nitrogen	0.14
Sulfur	0.03
Ash	0.60

178 Note: See Table S1 in SM for data references.

179

180 Feedstock particle size affects heat and mass transfer conditions. In general, the smaller particle  
 181 size of feedstock contributes to higher syngas yield and conversion efficiency (Parthasarathy and  
 182 Narayanan, 2014). However, an increased portion of particles with a size less than 1 mm results in  
 183 less hydrogen in the product gas, while the other gases (CO and CH<sub>4</sub>) are more along with  
 184 increased tar concentration in DFBR (Wilk and Hofbauer, 2013). Fluidized bed gasifiers can  
 185 handle fuels with particle diameters varying between 0.1 and 20 mm (Wood and Branch, 1986).  
 186 The biomass particle size in this study is assumed to be less than 2 mm after biomass preparation  
 187 for favorable conditions for product yields, process efficiency, and energy consumption (Andre et  
 188 al., 2020; García-Labiano et al., 2016; Sansaniwal et al., 2017; Shahbaz et al., 2021).

189 *2.1.2. Key modeling assumptions and methods*

190 Process modeling and simulation have served as a powerful tool for analyzing gasification  
 191 technology (Meramo-Hurtado et al., 2020). The plant scale in the process model in this study is  
 192 assumed to be 1,500 dry short tons/day (1,361 dry metric tons/day) of feedstock. Different scales  
 193 were explored to understand the impacts of scales (from 100 to 5,000 dry short tons/day). This  
 194 study used the thermodynamic property package Peng-Robinson with the Boston-Mathias  
 195 modifications (PR-BM) in Aspen Plus models, which have been widely recommended for high-  
 196 pressure hydrocarbon applications such as gas-processing, refinery, and petrochemical processes  
 197 (Gonzalez-diaz et al., 2021; Huang and Jin, 2019; Zhang et al., 2021). The process model employs  
 198 different property methods to represent the thermodynamics associated with each process section.

199 For example, the “SOLIDS” property method (Aspen Technology Inc., 2001) is used for biomass  
200 size reduction since it is designed for solids processing, where biomass and ash were specified as  
201 non-conventional components. The HCOALGEN and DCOALGEN models (Aspen Technology  
202 Inc., 2001) were used for calculating the enthalpy and density of the solids, respectively. The CC  
203 section uses “ELECNRTL” to handle molecular interactions for electrolyte solutions where  
204 monoethanolamine (MEA) is the solvent, this method was chosen given its capability to handle  
205 mixed solvent systems at any concentration (Aspen Technology Inc., 2001). The “Peng-Robinson”  
206 model was used for the syngas cleaning and air separation sections, and this method uses advanced  
207 alpha function and asymmetric mixing rules to accurately model polar, non-ideal chemical systems  
208 (Bisotti et al., 2021; Yu et al., 2021). An overview of the units and operating conditions in each  
209 section is presented as in SM Section S2 Units and Operating Conditions.

210 *2.1.3. Scenario analysis*

211 Separating CO<sub>2</sub> from different gas streams requires additional energy and expenditure, and  
212 generates environmental footprints. At the same time, the energy supply and fuel options for  
213 hydrogen plants have direct impacts on CO<sub>2</sub> quantity and sources (biomass versus fossil fuels). To  
214 investigate these complex interactions in the poly-generation system, the scenario analysis  
215 emphasizes CC implementation and energy supply options, as well as evaluates economic metrics  
216 and environmental impacts of different scenarios. The results of the scenario analysis will  
217 contribute to a better understanding of implementing heat and power supply strategies and the  
218 choice of CC. Table 2 outlines the scenario analysis settings adopted in this study.

219

220 **Table 2.** Scenario analysis settings.

	Scenario 1 Fully Electricity Self-Sufficient	Scenario 2 No CC	Scenario 3 Partially Electricity Self-Sufficient	Scenario 4 External Electricity
CC	Yes		Yes	Yes
CHP	Yes	Yes	Yes	
Combusting partial syngas for energy self-sufficiency	Yes			

221 Note: CC: Carbon Capture. CHP: Combined Heat and Power. Energy self-sufficiency: The  
222 biorefinery fulfills its energy requirement.

223 Scenario 1 includes CC and CHP, and combusts partial syngas in CHP to reach electricity self-  
224 sufficiency. As electricity is a valuable co-product that is often explored in previous TEA and LCA  
225 for biomass-based systems (Echeverria et al., 2021; Lan et al., 2021a), 30% of syngas  
226 (International Energy Agency Greenhouse gas R&D Programme, 2008) was modeled in Scenario  
227 1 that not only meets the internal electricity demand but also provides electricity surplus. Surplus  
228 electricity can be sold to the grid and bring additional revenue. For Scenario 2, the hydrogen  
229 production system does not consider CC, which is a baseline to understand the impacts of  
230 implementing CC in other scenarios. A CHP is deployed to burn the off gas from PSA (e.g.,  
231 containing CO, CH<sub>4</sub>, and remaining H<sub>2</sub>) to reduce the overall system energy requirement. Grid  
232 electricity from US West is imported externally if the electricity supply is not sufficient. When

233 surplus electricity is produced (Scenario 1), it is assumed to substitute grid electricity production  
234 mix from US West (WECC). Similar to Scenario 1, Scenario 3 considers CC technology to  
235 separate CO<sub>2</sub> from different CO<sub>2</sub> sources, including syngas, gasification off-gas, syngas cleaning  
236 off-gas, and CHP flue gas (as discussed in S2 of SM). Scenario 3 uses CHP to combust the off gas  
237 from PSA, representing a partially energy self-sufficient case (as the electricity generated from  
238 CHP is not sufficient to meet all internal electricity demand). Different from Scenario 1, all the  
239 syngas product is used for hydrogen production in scenario 3. Scenario 4 adopts CC technology  
240 but does not deploy the CHP plant. Instead, a combustor is used to recover heat from PSA off-gas  
241 and generate steam. All electricity demand in Scenario 4 is met by external CHP plants. Scenario  
242 4 represents the least energy self-sufficient scenario.

243 *2.2. Techno-economic analysis*

244 The mass and energy balance data from the Aspen model were used to size, map the equipment,  
245 and build the capital and operating cost profiles. Additionally, the capital costs of gasifiers were  
246 collected from the literature (data in Table S2 of SM). Once the capital and operating costs were  
247 determined, a discounted cash flow rate of return analysis was conducted to calculate the minimum  
248 selling price (MSP) of hydrogen. The minimum selling price (MSP) corresponds to the product  
249 selling price that makes net present value (NPV) equal to zero, considering all cash inflows and  
250 outflows from capital repayments, operation and maintenance, revenues, income tax rates and tax  
251 reductions due to plant depreciation (Nguyen and Clausen, 2019). MSP is widely used by the U.S.  
252 Department of Energy (DOE) for funding decisions related to biofuels (U.S. DOE, 2022) and  
253 establishing technical targets for hydrogen technology development (U.S. DOE, 2020b).

254

255 *2.2.1. Financial assumptions*

256 Table 3 shows the financial assumptions used in this study, which are consistent with the  
257 previous process simulation studies by the U.S. national laboratories (Humbird et al., 2011; Jones  
258 et al., 2013; Spath and Mann, 2004). The chemical/material/energy prices are documented in Table  
259 S3 in SM. The prices are adjusted to 2018 USD using the Producer Price Index (PPI) (US bureau  
260 of labor statistics, 2022).

261

262 **Table 3.** Parameters for the discounted cash flow analysis.

Parameters	Value/assumptions	References
Location	U.S.	
Plant life	30 years	(Spath and Ringer, 2005)
Year of analysis	2018	
Plant capacity	1500 dry short tons of feedstock/day	
Operating hours	8410 hrs/year	(Humbird et al., 2011)
Discount rate	10%	(Spath and Ringer, 2005)
Federal tax rate	21%	(IRS, 2022)
Depreciation method	USA IRS Modified Accelerated Cost Recovery System (MACRS)	(Humbird et al., 2011)
Depreciation Period (Years)		(Humbird et al., 2011)

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General Plant	7	
Steam/Electricity System	20	
Equity	40%	(Jones et al., 2013)
Loan Interest	8%	(Jones et al., 2013)
Loan Term, years	10	(Jones et al., 2013)
Working Capital (% of FCI*)	5%	(Humbird et al., 2011)
Salvage Value		(Humbird et al., 2011)
General Plant	0	
CHP Plant	0	
Construction Period (Years)	3	(Spath and Ringer, 2005)
% Spent in Year -2	8%	
% Spent in Year -1	60%	
% Spent in Year 0	32%	
Start-up Time (Months)	6	(Spath and Ringer, 2005)

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\*FCI is the total fixed capital investment, which is the sum of direct and indirect capital costs.

263

## 264 2.2.2. Cost metrics

265 This study includes several cost metrics, including CAPEX, OPEX, MSP of H<sub>2</sub>, and CC cost.  
 266 BECCS often produces multiple products, including hydrogen, carbon dioxide, electricity, and  
 267 other products (char and hydrogen sulfide). Determining the cost metrics for a multi-product  
 268 system is complicated as the production cost of one product is affected by the revenue of selling  
 269 other products made in the same system. Therefore, in this study, we first analyzed CAPEX and  
 270 OPEX for the entire BECCS biorefinery without distinguishing the cost of individual products,  
 271 then the MSPs of H<sub>2</sub> and CO<sub>2</sub> were quantified to explicitly explore the hydrogen and carbon  
 272 economics and their interactions with each other.

273 As a by-product of BECCS, the CO<sub>2</sub> price needs to be determined when estimating the MSP  
 274 of hydrogen. Carbon prices have different types, such as an emission trading system, carbon tax,  
 275 and carbon offset (The world bank, 2022). For example, in the U.S., the sequestration tax credit  
 276 45Q provides tax credits for carbon captured and sequestered, and the credit amount depends on  
 277 the type of project (Congressional Research Service, 2021). Different carbon prices have been  
 278 reported globally, depending on the policy and specific carbon programs. With this complexity in  
 279 mind, this TEA study explored a range of carbon prices reported in the literature, which can help  
 280 inform business and investment decisions by evaluating the impact of carbon prices on their  
 281 operations, identifying potential revenue opportunities/risks, and testing the potential impact of  
 282 climate change policies on their investment portfolios. In addition to geological storage, high  
 283 purity CO<sub>2</sub> (more than 99.5%) can be sold as an industrial gas, although geological storage is more  
 284 climate favorable and contributes to net carbon removal. Different CC and utilization pathways  
 285 have been explored in previous studies, therefore not included in this study (Zimmermann et al.,  
 286 2020).

287 2.3. Life cycle assessment

288 2.3.1 Goal and scope

289 We performed an ISO 14040 standard series compliant, attributional LCA of forest residue  
290 gasification with and without CC (depending on the scenarios).

291 The functional unit is 1 kg H<sub>2</sub> at a pressure of 30 atm with a purity higher than 99.9%. H<sub>2</sub> is  
292 usually the determining product given its mature market, choosing 1 kg H<sub>2</sub> as the functional unit  
293 allows for benchmarking and cross-reference comparisons with previous literature (Salkuyeh et  
294 al., 2018; Susmozas et al., 2016). To better understand the functionality of CO<sub>2</sub> removal, an  
295 additional functional unit of 1 kg CO<sub>2</sub> captured was included, allowing future studies for  
296 investigating different carbon negative technologies. The system boundary is cradle-to-gate,  
297 including raw material acquisition, transportation, and hydrogen production (see Fig. 1).

298 2.3.2. Inventory analysis

299 The LCI data of background processes were mainly obtained from ecoinvent database v3.6,  
300 unit model “allocation, cut-off by classification” (Wernet, et al., 2016), while the forest residue  
301 preprocessing and transportation data are from USLCI (National Renewable Energy Laboratory,  
302 2012). The preprocessing includes collection, chipping (to improve transportation efficiency), and  
303 field drying. The transportation mode is a combination truck powered by diesel, the transportation  
304 distance is 68 km that covers the steps from the collection site to the regional storehouse and from  
305 the regional storehouse to the conversion facility. The LCI data of the foreground process (e.g.,  
306 gasification) are from process simulations discussed above, and have been normalized based on 1  
307 kg of hydrogen (the functional unit). The forest residue used for gasification is a product of  
308 sustainable forestry of two species (Douglas fir and Ponderosa pine) grown in the Pacific  
309 Northwest U.S. The CO<sub>2</sub> captured are assumed to be geologically stored permanently, but the costs  
310 and environmental impacts of further transportation to geological sites and storage are not included  
311 in this study. The electricity co-product credits (in Scenario 1) were estimated based on the  
312 substitution of grid electricity production mix in the western U.S. (WECC). The system expansion  
313 is used by following ISO standard 14044 to avoid allocation wherever possible (International  
314 Organization for Standardization, 2006). Other products such as biochar and hydrogen sulfide are  
315 cut off due to less than 0.1% contribution to the mass of total product outputs.

316 2.3.3. Impact assessment

317 The TRACI 2.1 method (EPA, 2022) was used for life cycle impact assessment (LCIA). The  
318 environmental impact categories cover ozone depletion, global warming, acidification,  
319 eutrophication, smog formation, human health impacts, ecotoxicity, and fossil fuel depletion.

320

321 **3. Results and discussion**

322 Based on the simulation results (mass and energy balance) of the biomass gasification plant,  
323 the TEA and LCA results are reported in this section. The technical parameters of the process  
324 model are presented and compared to the literature in the first section for model validation.  
325 Subsequently, the economic and environmental performance for different scenarios are presented.  
326 In addition, the trade-off impacts of CC integration into hydrogen production are discussed.

327 3.1. Process model and result validation

328 The syngas composition result of the process simulation is: 40.6 mol% of hydrogen, 14.7 mol%  
329 of CO, 10.4 mol% of CO<sub>2</sub>, 34 mol% of water, and small amounts of other gases, which are  
330 consistent with the literature (Göransson et al., 2011; Pala et al., 2017). The mass and energy  
331 balance results are documented in Table S5 in SM. The carbon distribution is reported in Fig. S1  
332 in SM and used to calculate the carbon capture rate that reflects the fraction by which carbon  
333 emissions are captured relative to the total carbon inputs (Trinks et al., 2020). The carbon capture  
334 rate of this study is 87% calculated by dividing the amount of carbon captured by the total amount  
335 of carbon inputs, including carbon in biomass and lean MEA solvent (0.3 mol%). The carbon  
336 capture rate in this study is higher than the literature value 31%–60% (Fernanda Rojas Michaga et  
337 al., 2022; Salkuyeh et al., 2018; Susmozas et al., 2016) because previous studies only considered  
338 one or two carbon emission sources (e.g., boilers or syngas). In contrast, this study considers all  
339 carbon emission sources in the biorefinery, including gasification, syngas, gas cleaning, and CHP.  
340 The carbon capture rate for BECCS in this study is also comparable with direct air capture (e.g.,  
341 85.4% and 93.1% depending on the electricity source) (Deutz and Bardow, 2021).

342 3.2. TEA Results

343 3.2.1 Production cost profiles

344 Fig. 2 presents capital expenditure (CAPEX) and yearly operating expenditure (OPEX) for all  
345 scenarios. Fig. 2 (a) shows that the gasification section is the major contributor to CAPEX for all  
346 scenarios. This agrees with previous studies on biomass gasification for other products such as  
347 electricity/hydrocarbon or coal-biomass co-gasification systems (Arnaiz del Pozo et al., 2021b;  
348 Schweitzer et al., 2018; Wang et al., 2013). The second-largest contributor to CAPEX is ASU.  
349 Similar high CAPEX of ASU have been reported in the literature (AlNouss et al., 2020; Ebrahimi  
350 et al., 2015; Ebrahimi and Ziabasharhagh, 2017; Prakash Rao and Michael Muller, 2007; Young  
351 et al., 2021). ASU provides pure oxygen instead of air to the combustion process so that a higher  
352 CO<sub>2</sub> concentration in the flue gas is obtained, facilitating the following CC. Moreover, ASU  
353 provides nitrogen for the gas cleaning section, which benefits the entire system through service  
354 sharing. Removing ASU will reduce CAPEX but significantly increase OPEX, given the need to  
355 purchase nitrogen and oxygen. To quantitatively explore this impact, a comparison of the BECCS  
356 system with and without the ASU was made (Table S7 of SM). It shows that the absence of ASU  
357 can increase or decrease the MSP of H<sub>2</sub> depending on the trade-offs between the increased cost for  
358 purchased O<sub>2</sub> and N<sub>2</sub> and decreased electricity cost and CAPEX.

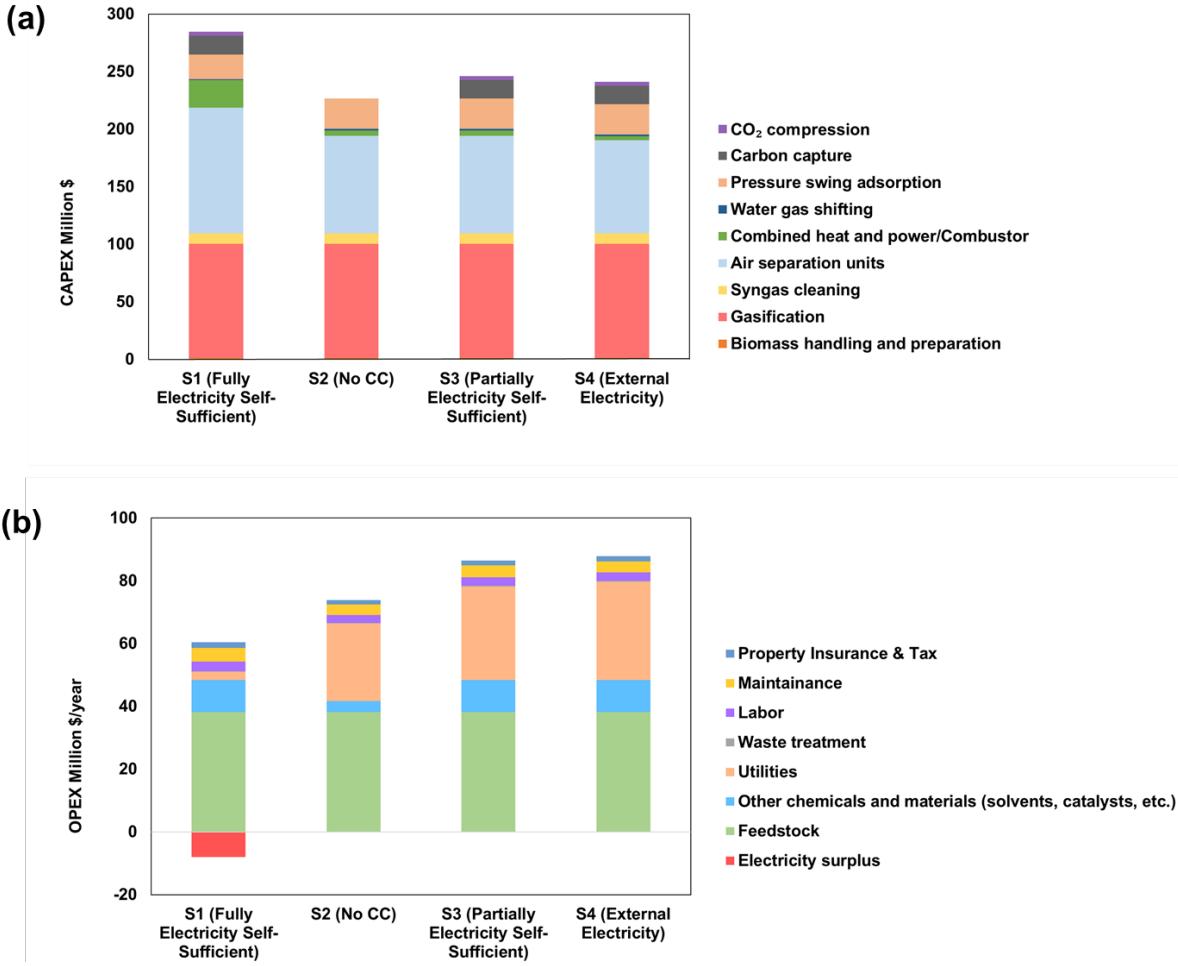
359 Across four scenarios, Scenario 1 (Fully Electricity Self-Sufficient) has the highest CAPEX  
360 due to the additional capital needed for electricity self-sufficiency. The CAPEX is reduced as the  
361 electricity self-sufficiency is decreased in Scenario 3 (partially self-sufficient, 13% reduction of  
362 CAPEX) and Scenario 4 (all externally purchased electricity, 15% reduction of CAPEX). The  
363 highest CAPEX of Scenario 1 (Fully Electricity Self-Sufficient) is attributed to CHP and ASU.  
364 This is due to the higher capacity of CHP and ASU, which burn syngas and provide more oxygen  
365 for burning syngas, respectively. The benefit of electricity self-sufficiency is reduced OPEX, as  
366 shown in Fig 2.(b). Scenario 2 (No CC) has the lowest CAPEX, given the absence of CC. The

367 incorporation of the CC section increases the total CAPEX by 9% (comparing Scenarios 2 and 3).  
368 Another CAPEX contributor is the PSA unit (8%–10% of the total CAPEX). The contribution of  
369 the rest of the operating units is minor. For Scenario 1, the annualized CAPEX (calculated using  
370 Equation S1 in SM) takes about 36% of the total hydrogen production cost.

371 Compared with CAPEX, OPEX results in Fig. 2(b) show different trends of scenarios. Scenario  
372 1 (Fully Electricity Self-Sufficient) has the lowest OPEX because of the lowest utilities achieved  
373 by the full electricity self-sufficiency. In contrast, Scenario 4 (External Electricity) has the highest  
374 OPEX caused by the highest utilities, most from electricity purchases (75%). Another 25% of  
375 utility costs are for heat/cooling energy. The main contributors to electricity consumption are ASU  
376 (71%), PSA (18%), CO<sub>2</sub> compression (8%), biomass preparation (2%) and CC (1%). Although CC  
377 does not consume much electricity, it is the major contributor to heat/cooling energy (51%),  
378 followed by ASU (26%) and gas cleaning (23%). This is why the inclusion of the CC section  
379 increases the utilities by 20%, comparing Scenario 3 (Partially Electricity Self-Sufficient) with  
380 Scenario 2 (No CC). Including CC also increase the usage of other raw materials such as solvent  
381 and water by 1.8 folds.

382 In addition to utilities, the major contributor to the OPEX is feedstock cost (forest residues).  
383 The high contribution of biomass feedstock is consistent with previous studies (Li et al., 2020;  
384 Wang et al., 2019), where the cost of biomass accounts for at least 30% of the total production  
385 cost. Given the large contribution of feedstock costs, forest residue price is included in the  
386 sensitivity analysis to understand the impacts of varying feedstock prices.

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**Fig. 2.** Results of CAPEX (a) and OPEX (b) breakdown for four scenarios.

392

### 3.2.2. MSP of Hydrogen and carbon price

393

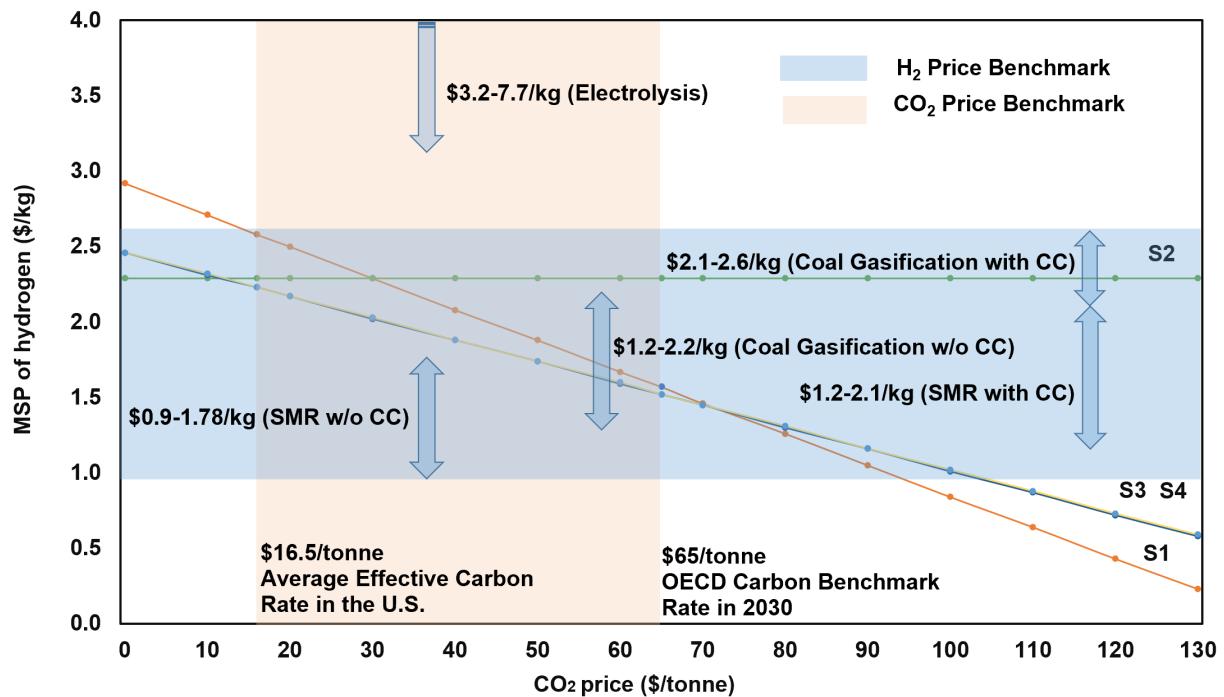
As discussed in Section 2.3, hydrogen cost depends on CO<sub>2</sub> price. Fig. 3 presents the effect of the CO<sub>2</sub> price on the hydrogen MSP in different scenarios. Scenario 2 (No CC) is a flat line as the exclusion of CC. Scenarios 3 (Partially Electricity Self-Sufficient) and 4 (External Electricity) are almost overlapped because of their similar CAPEX and OPEX (as demonstrated in Fig. 2). Blue areas represent benchmarked price ranges of H<sub>2</sub> made from fossil fuels with and without CC. The price of fossil-based hydrogen without CC ranges from \$0.9-1.78/kg H<sub>2</sub> (steam methane reforming, SMR) to \$1.2-2.2/kg H<sub>2</sub> (coal gasification) (IEA, 2020; National Research Council, 2004). When CC is included in SMR and coal gasification, their prices increase to \$1.2-2.6/kg H<sub>2</sub> (IEA, 2020; Parkinson et al., 2019). SMR and coal gasification were chosen as benchmark technologies because SMR contributes to 76% of the global H<sub>2</sub> production and coal gasification contributes to 22% (Lepage, et al. 2021). The carbon price benchmark (orange area in Fig. 3) uses the effective carbon rate of the U.S. reported by OECD (Organisation for Economic Co-operation and Development), which estimates an average carbon price from taxes and emission trading

405 systems in different countries (OECD, 2021). According to this study, the carbon price in the U.S.  
406 is \$16.5/tonne in 2021 and the OECD benchmarked rate for the U.S. in 2030 is projected to be  
407 \$65/tonne.

408 Fig. 3 leads to two conclusions. First, forest residue-derived H<sub>2</sub> is economically competitive  
409 with current fossil-based H<sub>2</sub> with CC. The MSP of H<sub>2</sub> ranges from \$1.52 – 2.92/kg H<sub>2</sub> when the  
410 carbon price is \$0–65/tonne of CO<sub>2</sub>. Scenarios 3 (Partially Electricity Self-Sufficient) and 4  
411 (External Electricity) have similar MSP to coal-based H<sub>2</sub> with CC at a price of \$0 – 19/tonne of  
412 CO<sub>2</sub>. With a carbon price higher than \$19/tonne of CO<sub>2</sub>, Scenarios 3 and 4 are more economically  
413 favorable than coal-based H<sub>2</sub> with CC. Scenario 1 (Fully Electricity Self-Sufficient) has a similar  
414 MSP with fossil-based systems (SMR and coal gasification) with CC at \$16.5 – 85/tonne of CO<sub>2</sub>.  
415 The benchmarked CO<sub>2</sub> price range (\$16.5 – 65/tonne of CO<sub>2</sub>) is within the CO<sub>2</sub> price range which  
416 makes Scenario 1, 3, and 4 economically feasible. When the CO<sub>2</sub> price is higher than \$89/tonne of  
417 CO<sub>2</sub>, all scenarios are more economically attractive than fossil H<sub>2</sub> with CC. Retrofitting CC from  
418 existing fossil-based facilities can be less feasible than newly built plants (Arasto et al., 2013), thus  
419 integrating CC into bio-based H<sub>2</sub> production can be practically attractive. Compared to coal  
420 gasification without CC, Scenarios 3 and 4 can be economically feasible with a CO<sub>2</sub> price higher  
421 than \$18.3/tonne. Scenario 1 needs a higher CO<sub>2</sub> price ( $\geq$ \$33/tonne of CO<sub>2</sub>) to achieve similar  
422 economic feasibility. Compared to SMR without CC, Scenario 1 needs a price higher than  
423 \$55/tonne of CO<sub>2</sub>, and Scenarios 3 and 4 need a price higher than \$47/tonne of CO<sub>2</sub>. These CO<sub>2</sub>  
424 prices are still within the benchmarked CO<sub>2</sub> price range (\$16.5 – 65/tonne of CO<sub>2</sub>).

425 The second conclusion is that CO<sub>2</sub> prices determine the comparative economic competitiveness  
426 of three scenarios with CC. Scenario 1 (Fully Electricity Self-Sufficient) is less economically  
427 favorable when CO<sub>2</sub> price is low but more attractive when CO<sub>2</sub> price is higher than \$70/tonne of  
428 CO<sub>2</sub>. The lower production rate of H<sub>2</sub> can explain this in Scenario 1, where syngas is combusted  
429 to achieve electricity self-sufficiency and thus a higher CO<sub>2</sub> price is needed to compensate H<sub>2</sub> loss  
430 (see Table S4 in SM for the production rate of H<sub>2</sub> and CO<sub>2</sub> in four scenarios). The incorporation  
431 of CC increases the hydrogen MSP by 7% by comparing Scenarios 2 (No CC) and 3 (Partially  
432 Electricity Self-Sufficient) at a CO<sub>2</sub> price of \$0/kg. However, revenue from CO<sub>2</sub> more than fully  
433 offsets the additional cost of CC when the carbon price is greater than \$12/tonne. Fig. 3 also  
434 includes a benchmark for electrolyzed H<sub>2</sub> with a much higher price (~\$3.2-7.7/kg H<sub>2</sub>) (IEA, 2020;  
435 IRENA, 2020). Forest residue-derived H<sub>2</sub> is much more economically feasible based on the MSP  
436 shown in Fig. 3, compared to the current cost of electrolyzed H<sub>2</sub>.

437



**Fig. 3.** Effect of the carbon price on the Minimum Selling Price (MSP) of hydrogen.

#### 442 3.2.3. Sensitivity analysis

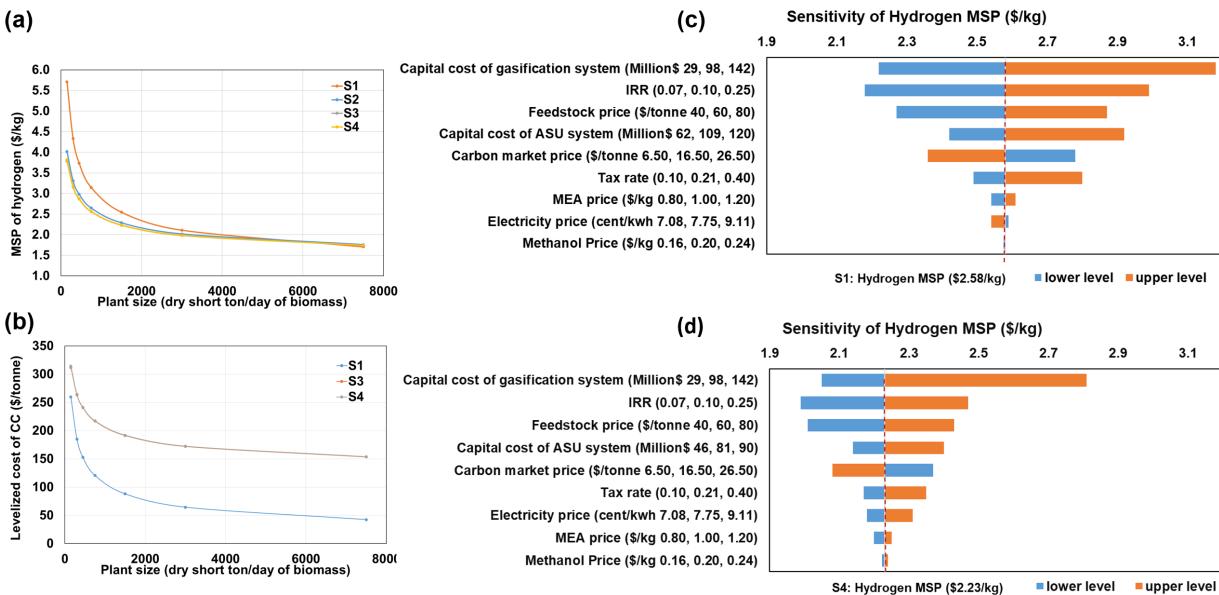
443 The sensitivity analysis focuses on understanding the effects of plant sizes and financial  
 444 parameters with variations in this TEA. These parameters include financial assumptions that  
 445 significantly impact the economic feasibility of biomass conversion technologies in general (Lan  
 446 et al., 2021a), and prices of material and energy inputs, as listed in Table S3 in SM.

447 Fig. 4 (a-b) shows the hydrogen MSP and levelized cost of cost when the plant size varies from  
 448 150-7500 dry short ton/day of biomass for all scenarios. The levelized cost of CC was estimated  
 449 as detailed in Section S7 of SM (Keith et al., 2018), which is the cost required for building and  
 450 operating the CC units to the physical amount of CO<sub>2</sub> captured from the given point of hydrogen  
 451 plant (IEA, 2021; Roussanaly, 2019; Roussanaly et al., 2021). The CAPEX of different plant sizes  
 452 was estimated using a scaling factor of 0.6, the most commonly used value for chemical  
 453 engineering unit operations (Tribe and Alpine, 1986). The OPEX components such as materials  
 454 cost, waste streams, utilities, maintenance (OPEX except for labor), and production rate have been  
 455 assumed proportional to the size of the plant and linearly adjusted based on the plant capacity. The  
 456 labor cost was re-calculated for each case by using the empirical relationship between labor and  
 457 plant capacity, process section number and operating hours of the plant (Peters, et al., 2003)  
 458 (documented in Section 5 in SM). Fig. 4 (a) shows the MSP of hydrogen as a function of the plant  
 459 capacity, where the carbon price is fixed at \$16.5/tonne. The slope for plant's capacities between  
 460 150 and 2000 dry short ton/day is steep, resulting in a significant decrease in the MSP of hydrogen.  
 461 For a plant size beyond 2,000 dry short ton/day, the MSP of hydrogen continues to drop but at a

462 slower rate. On the other hand, increased size requires more biomass feedstock, which may be  
 463 limited in some regions. In conclusion, the results show that it may not be optimal to build such  
 464 BECCS biorefineries larger than 2,000 dry short ton/day from an economies of scale point of view.

465 Similar trends are observed in Fig. 4(b) where hydrogen price is fixed at the market price of  
 466 \$1.26/kg (SMR w/o CC). Economies of scale have a more significant impact on CC cost at smaller  
 467 sizes (150 and 1,000 dry ton/day), as shown in Fig. 4 (b). Although CC has a relatively low  
 468 contribution (9%) to the total CAPEX, CO<sub>2</sub> is a primary product whose production increases in a  
 469 greater proportion than the increase in its cost at smaller sizes. The comparisons among four  
 470 scenarios show different trends in Fig. 4 (a-b). In Fig. 4(a), Scenario 1 (Fully Electricity Self-  
 471 Sufficient) has the highest hydrogen MSP, and the differences between Scenario 1 and other  
 472 scenarios are diminished as plant size increases. However, the opposite trend is observed in Fig.  
 473 4(b), where Scenario 1 shows a lower leveledized cost of CC than Scenario 3 (Partially Electricity  
 474 Self-Sufficient) and 4 (External Electricity) (Scenario 2 is not included due to the exclusion of  
 475 CC), and the differences between Scenario 1 and others increase as plant sizes increases. The  
 476 different trends in Fig. 4 (a) and (b) can be explained by different product focuses. When the  
 477 product focus is H<sub>2</sub>, Scenario 1 is less favorable due to lower H<sub>2</sub> production (Table S4 in SM); on  
 478 the contrary, when the product focus is CO<sub>2</sub>, scenario 1 is more favorable given lower utility costs  
 479 (as demonstrated in Fig. 2.b).

480



481

482

483 **Fig. 4.** Effect of plant size on the MSP of hydrogen (a) and the leveledized cost of CC (b);  
 484 Sensitivity (considering maximum and minimum values) of key parameters in Scenario 1 (Fully  
 485 Electricity Self-Sufficient) (c) and Scenario 4 (External Electricity) (d). References for the  
 486 uncertainty range of parameters are in Table S2 and Table S9 in SM.  
 487

488 Results from the sensitivity analysis of key parameters are presented in Fig. 4 (c) and (d) for  
489 the two extreme cases – Scenario 1 (electricity self-sufficient) and Scenario 4 (all electricity  
490 externally supplied). The uncertainty range of each parameter is based on the data points collected  
491 from the literature (Table S2 for gasification price data, Table S9 for other parameters). Only  
492 parameters with significant impacts on the results are shown (lead to > 0.1% variation of the  
493 results). The two scenarios have similar ranks for most parameters except electricity price that has  
494 opposite effects in Scenarios 1 and 4. Electricity price affects the revenue of selling surplus  
495 electricity in Scenario 1, therefore increasing electricity price decreases hydrogen MSP as shown  
496 in Fig. 4 (c). As electricity is purchased externally in Scenario 4, increasing electricity price raises  
497 hydrogen MSP, as demonstrated by Fig. 4(d). When more electricity is internally produced  
498 (Scenario 1), the biorefinery is more resilient to the electricity price fluctuations, although internal  
499 electricity production has higher hydrogen MSP at fixed and moderate electricity prices than other  
500 scenarios (discussed in Section 3.2.2 for Fig. 3.). It also indicates that hydrogen economics of  
501 Scenario 4 will likely be more sensitive to renewable energy access given its high sensitivity to  
502 electricity price. The gasification system can be of the greatest uncertainty due to the wide range  
503 of gasifier cost estimates from different literature (Table S2 in SM). The MSP of hydrogen is also  
504 sensitive to feedstock price, which is influenced by regional supply and demand, e.g., demand for  
505 alternative uses of forest residues such as for electricity and fuel production (Daioglou et al., 2016).  
506 The risk of volatilities in the feedstock price can be limited by developing partnerships with  
507 biomass suppliers (e.g., forest management corporations, communities) and establishing reliable  
508 logistic infrastructure for a steady cost. Following feedstock price, the MSP of hydrogen is also  
509 sensitive to CAPEX of ASU. With technology improvement, CAPEX could decrease and improve  
510 the economic feasibility of hydrogen. The substantial impacts of CO<sub>2</sub> prices have already been  
511 demonstrated in the previous section when CO<sub>2</sub> prices have large variations. Finally, the IRR  
512 (discount rate) has a significant impact on the MSP of hydrogen, this is due to the contribution of  
513 CAPEX, which directly connects to profitability. The tax rate and prices of chemicals such as  
514 MEA and methanol have minor influences on the MSP of hydrogen.

### 515 3.3. LCA Results

#### 516 3.3.1. Life cycle impact assessment results of hydrogen production pathways

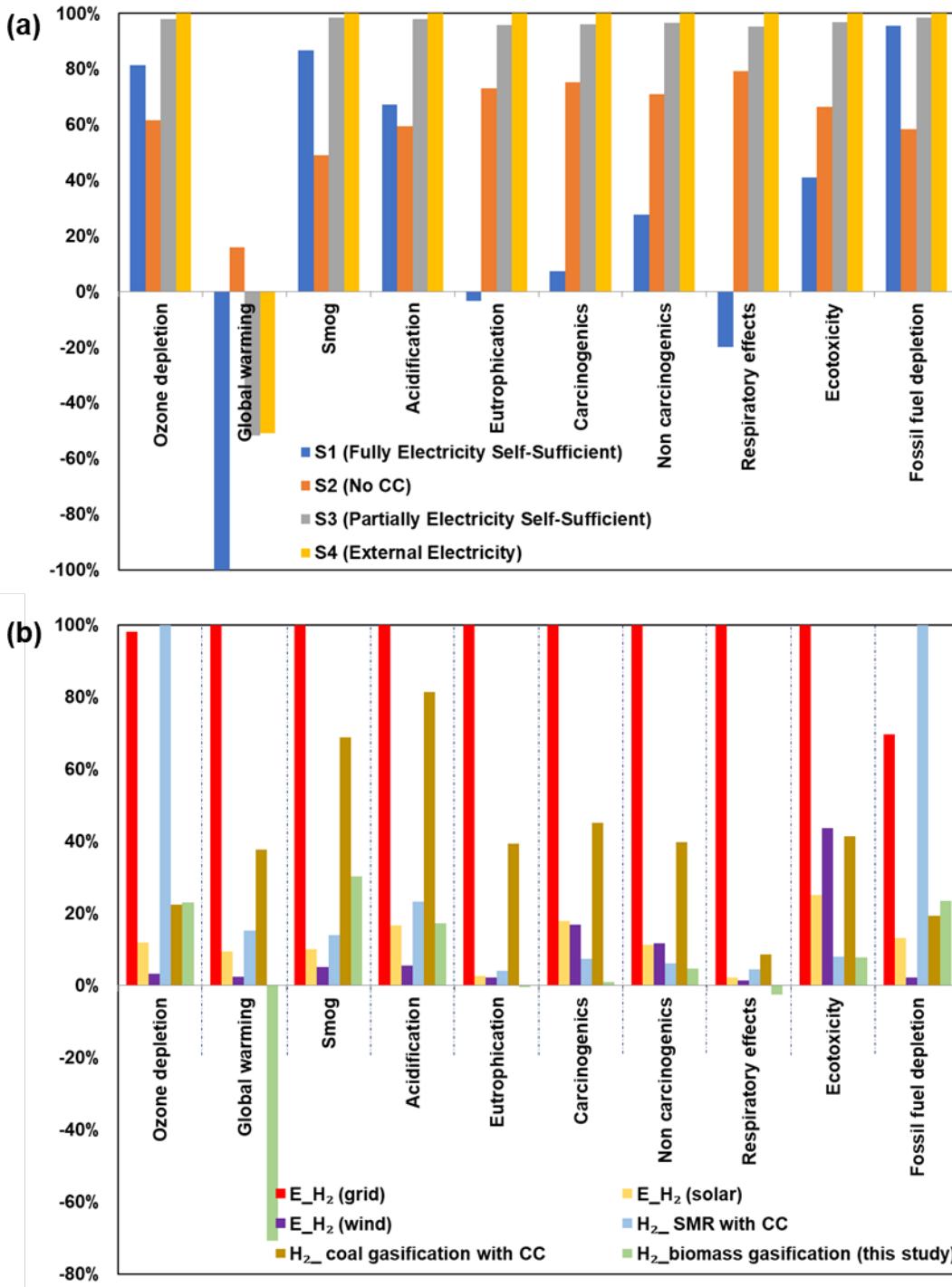
517 Fig. 5 presents the LCA results of four scenarios of biomass gasification and the comparison  
518 with alternative hydrogen pathways (hydrogen from electrolysis and fossil fuels with CC) based  
519 on the functional unit of 1 kg of H<sub>2</sub>. The LCA results of scenarios with 1 kg of CO<sub>2</sub> as the functional  
520 unit are shown in SM Fig. S2. Across four scenarios in Fig 5 (a), Scenario 1 has the lowest  
521 environmental impacts on GWP, eutrophication, human health impacts (including carcinogenics  
522 and non-carcinogenics, and respiratory effects), and ecotoxicity. LCA results with 1 kg of CO<sub>2</sub> show  
523 similar trends (see SM Fig. S2). For the other environmental impact categories (i.e., ozone  
524 depletion, smog, acidification, and fossil fuel depletion), Scenario 2 without CC is the lowest.  
525 Excluding Scenario 1, Scenario 2 has the lowest results across all environmental impact categories  
526 compared to Scenario 3 and 4. The higher environmental impacts of Scenarios 3 and 4 are caused  
527 by increased energy and chemical (e.g., solvent) consumption, which are further discussed in  
528 Section 3.3.2. This observation concludes that implementing CC in gasification increases

529 environmental impacts in general, however, some increased impacts can be substantially mitigated  
530 by self-sufficient energy design.

531 As Scenario 4 relies on imported electricity, environmental impacts could be much lower in  
532 the future if the electricity grid is deeply decarbonized or have a high degree of energy generated  
533 from renewable and low carbon sources. To explore the impact of renewable energy access, two  
534 additional cases were analyzed for Scenarios 1 and 4 by changing the electricity source from the  
535 current grid to solar and wind. The results are shown in SM Fig. S5 and S6. The results show that  
536 the electricity self-sufficiency design in Scenario 1 is no longer preferable when the biorefinery  
537 has access to solar and wind in Scenario 4. These results highlight the need to consider renewable  
538 energy access in biorefinery energy design. Future research can investigate the impacts of different  
539 grid decarbonization scenarios (Phadke et al., 2020; United States Department of State, 2021) on  
540 the optimal energy design of BECCS.

541 The life cycle GWP of Scenario 2 (No CC) is 2.99 kg CO<sub>2</sub> equivalent per kg of hydrogen  
542 produced (see LCIA results in Table S6). The life cycle GWP results in other scenarios are negative  
543 (-18.8, -9.71 and -9.56 kg CO<sub>2</sub> equivalent per kg of hydrogen produced for S1, S3 and S4,  
544 respectively), which are attributed to carbon sequestered by biomass and captured by CC.  
545 Scenario 1 has more negative GWP than Scenario 3 and 4 since electricity is self-sustained instead  
546 of importing from the grid and CO<sub>2</sub> emissions from internal electricity generation in Scenario 1  
547 are also captured. This result highlights the climate benefits of self-sufficient energy design for  
548 forest residue-based BECCS.

549



551 **Fig. 5.** Comparisons of LCA results of 1 kg of hydrogen (a) from biomass gasification in four  
552 scenarios. (b) from biomass gasification in Scenario 1, electrolysis (E represents electrolysis),  
553 and fossil-based routes.

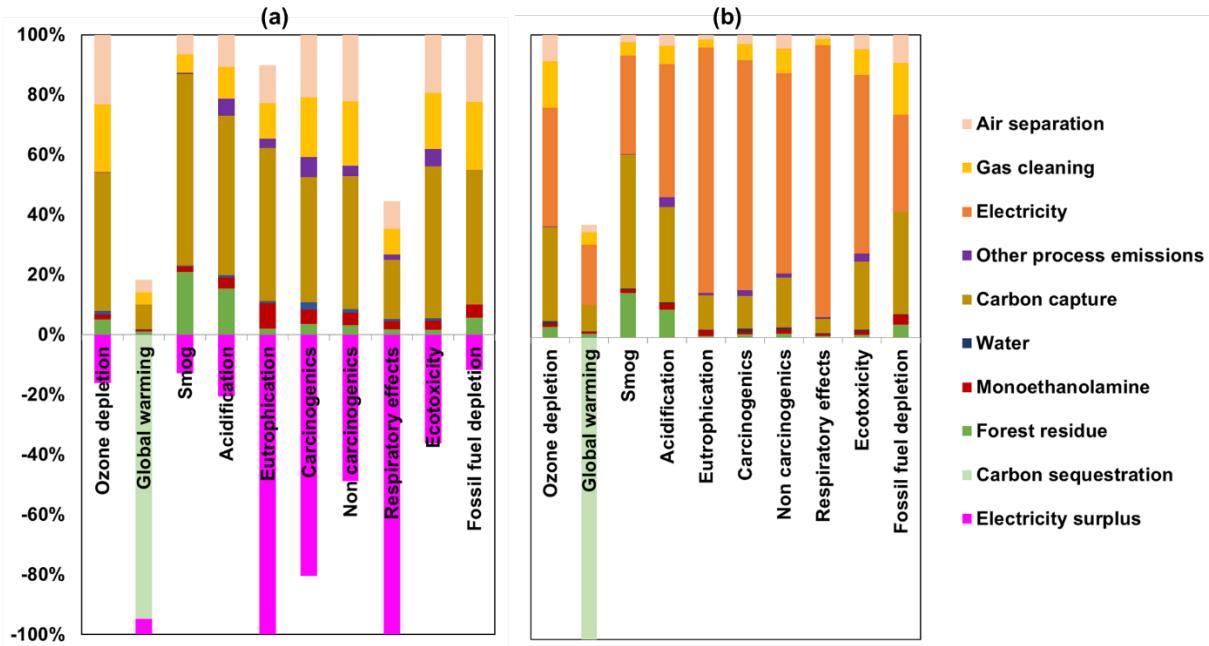
554 Currently, hydrogen can be made from different sources, including SMR, coal gasification,  
555 and electrolysis from different electricity sources (grid, solar, and wind). In Fig. 5 (b), the life cycle  
556 environmental profiles of different hydrogen production alternatives based on harmonized

557 literature data (Al-Qahtani et al., 2021) are compared at the same functional unit, 1 kg of hydrogen.  
558 The result shows that forest residue-derived H<sub>2</sub> has the greatest decarbonization potential  
559 compared to other alternative technologies. BECCS is the only pathway that provides carbon-  
560 negative hydrogen. Compared to fossil-based H<sub>2</sub>, forest residue-derived H<sub>2</sub> has the lowest impacts  
561 across all categories, except ozone depletion, smog, and fossil fuel depletion, where BECCS has  
562 slightly higher results than SMR/coal gasification with CC, indicating the co-benefits of BECCS  
563 in decarbonizing and reducing environmental impacts of current hydrogen production. Compared  
564 to H<sub>2</sub> relying on grid electricity, forest residue-derived H<sub>2</sub> has lower impacts across all categories.  
565 Compared with H<sub>2</sub> using wind and solar, BECCS has advantages in GWP, eutrophication,  
566 ecotoxicity, and human health impacts, including carcinogenics, non-carcinogenics, and  
567 respiratory effects. However, these advantages do not hold for the other environmental impact  
568 categories (ozone depletion, smog, acidification, and fossil fuel depletion). Note that electrolyzed  
569 H<sub>2</sub> is much more expensive (3.2-7.7\$/kg H<sub>2</sub>) than other options, and the significant cost reduction  
570 needs to come from electrolyzer and electricity (IRENA, 2020), which are challenging to achieve  
571 in the near term. Therefore, BECCS is still a promising option for clean H<sub>2</sub> in the near term.

572 *3.3.2. Contribution analysis*

573 Contribution analysis of life cycle environmental impacts is presented in Fig. 6 (a) and (b) for  
574 the two extreme cases – Scenario 1 (electricity self-sufficient) and Scenario 4 (all electricity  
575 externally supplied), respectively. CC is the most significant contributor to all impacts in Scenario  
576 1 due to its direct emissions (e.g., the solvent MEA evaporation, wastewater generation) and  
577 intensive energy and solvent consumption. For Scenario 4, electricity is the largest contributor,  
578 and this contribution can be reduced using renewable energy such as solar and wind (SM Fig. S3  
579 and S4). Other than CC and electricity, gas cleaning and air separation make significant  
580 contributions to the results in both scenarios. The impacts of biomass feedstock production (shown  
581 as “Forest residue” in Fig. 6) are two-fold. On the one hand, it accounts for a favorable  
582 decarbonization contribution (i.e., negative percentage) due to CO<sub>2</sub> sequestration during biomass  
583 growth. On the other hand, biomass production has a relatively high contribution to smog  
584 formation and acidification that can be caused by machinery energy consumption (e.g., diesel) at  
585 the landing system where the forest residues are preprocessed (e.g., chipping) and logging residue  
586 are extracted (Ranius et al., 2018).

587  
588  
589



591 **Fig. 6.** Contribution analysis for the life cycle environmental impacts of S1 Fully Electricity  
592 Self-Sufficient (a) and S4 External Electricity (b).

593 **3.4. Trade-offs and Co-Benefits between economics and environmental impacts**

595 The different options for designing biomass gasification as a BECCS pathway result in trade-  
596 offs and co-benefits between economics and environmental impacts. Incorporating CC into  
597 biomass gasification increases environmental impacts, CAPEX, and OPEX; however, it improves  
598 H<sub>2</sub> economics, depending on the CO<sub>2</sub> prices. Energy self-sufficiency allows for maximum  
599 decarbonization and mitigates the increase in environmental burdens of the BECCS system caused  
600 by the CC section. It also reduces CC costs. These are co-benefits of energy self-sufficiency in  
601 carbon economics and environmental impact. However, energy self-sufficiency worsens H<sub>2</sub>  
602 economics due to the reduced H<sub>2</sub> production, and this is a trade-off. Decision-makers could use  
603 these results to support the economic mechanisms for shared investments in CC and hydrogen  
604 production, considering carbon price, hydrogen and electricity market, and environmental  
605 regulations.

606 More recently in 2022, the U.S. government published the Inflation Reduction Act of 2022  
607 that presents the modified credit for carbon dioxide capture in section 45Q and credit for producing  
608 clean hydrogen in section 45V, although two credits cannot be used simultaneously (The U.S.  
609 Government Publishing office, 2022). Based on 45Q, carbon credits can be as high as \$85/ton CO<sub>2</sub>  
610 which can bring the MSP of H<sub>2</sub> to \$1.2/kg in S1, \$1.4/kg in S3 and S4 as shown in Fig. 3 (estimated  
611 in 2018 \$). Based on 45Q future section in 2023, H<sub>2</sub> credits can be up to \$3.00 for life cycle GHG  
612 emissions less than 0.45 kg CO<sub>2e</sub>/kg H<sub>2</sub>, \$1.00 for 0.45–1.5 kg CO<sub>2e</sub>/kg H<sub>2</sub>, \$0.75 for 1.5–2.5 kg  
613 CO<sub>2e</sub>/kg H<sub>2</sub>, and \$0.60 for 2.5–4.0 kg CO<sub>2e</sub>/kg H<sub>2</sub> (The U.S. Government Publishing office, 2022).  
614 Hence, in this study, the potential credit is \$3.00 for S1, S3, and S4 (life-cycle GWP for S1, S3,

615 and S4 is -18.8, -9.71, -9.56 kg CO<sub>2e</sub>/kg H<sub>2</sub>, respectively, see Section 3.3.1), and is \$0.60 for S2  
616 (no CCS) (life-cycle GWP 2.99 kg CO<sub>2e</sub>/kg H<sub>2</sub>). In this way, even without CO<sub>2</sub> credit (CO<sub>2</sub> price  
617 equals \$0 in Fig. 3), the MSP of hydrogen can be profitable. Hence, in the future, with 45V credit,  
618 hydrogen production with BECCS has huge potential economic advantage over current fossil fuel-  
619 based hydrogen. This also highlights the importance of biomass gasification hydrogen plant to  
620 meet the life-cycle GWP requirement of 0.45 kg CO<sub>2e</sub>/kg H<sub>2</sub> for securing hydrogen credits.

621

#### 622 4. Conclusion

623 This study evaluated the techno-economic feasibility and environmental impacts of  
624 gasification-based BECCS using forest residues. Different scenarios of incorporating CC and  
625 energy supply strategies were investigated and compared. Specifically, this study used the  
626 integrated TEES framework to quantify the economic and environmental impacts of such a  
627 biorefinery for its full decarbonization potential at the process level. The production cost profiles  
628 (CAPEX and OPEX) were built based on the mass and energy balance results from process  
629 simulations. While it is evident that the gasifier and ASU are the major CAPEX contributors, the  
630 inclusion/exclusion of ASU has different impacts on CAPEX and OPEX given its complex  
631 interactions with other process units in the biorefinery, highlighting the need for future research  
632 focusing on integrated system design instead of separated components.

633 CC incorporation increases the CAPEX and decreases the OPEX, yet resulting in an overall  
634 higher cost of hydrogen under current carbon prices. However, with increasing interest in  
635 decarbonization, CC incorporation is attractive and economically competitive with fossil-based  
636 routes with CC. Electricity supply is a crucial factor in determining OPEX. When electricity self-  
637 sufficiency is fully reached, the OPEX is lower than other cases. However, this benefit is not  
638 sufficient to fully offset CAPEX contribution to the overall economics when the carbon price is  
639 lower than \$70/tonne of CO<sub>2</sub>.

640 The economic competitiveness of forest residue-derived H<sub>2</sub> depends on CO<sub>2</sub> prices and H<sub>2</sub>  
641 price benchmarks. For example, the MSP of H<sub>2</sub> are \$1.52 – 2.92/kg H<sub>2</sub> with carbon prices of \$0–  
642 65/tonne of CO<sub>2</sub>. Compared to fossil-based H<sub>2</sub> with CC, BECCS with a self-sufficient electricity  
643 supply provides competitive H<sub>2</sub> at price higher than \$16.5/tonne of CO<sub>2</sub>; while BECCS with partial  
644 or no internal electricity supply has similar MSP of H<sub>2</sub> with coal gasification at low carbon prices  
645 \$0–19/tonne of CO<sub>2</sub> and MSP of H<sub>2</sub> with SMR at higher carbon prices (\$19–89/tonne of CO<sub>2</sub>).  
646 Compared to fossil-based routes without CC, higher CO<sub>2</sub> prices are needed for BECCS to be  
647 economically competitive, but these prices are still within the benchmarked CO<sub>2</sub> price range (\$16.5  
648 – 65/tonne of CO<sub>2</sub>). In a conclusion, with the benchmarked CO<sub>2</sub> prices, forest residue-derived H<sub>2</sub>  
649 is economically competitive compared with fossil-based H<sub>2</sub>.

650 The H<sub>2</sub> from BECCS is more cost-effective than current electrolyzed H<sub>2</sub> regardless of CO<sub>2</sub>  
651 prices. The sensitivity analysis shows that a plant size of 2,000 dry short ton/day can be the upper  
652 threshold to take advantage of the economies of scale. Establishing a reliable logistic infrastructure  
653 of feedstock supply is essential since hydrogen MSP is highly sensitive to forest residue price.

654 The LCA shows that implementing CC in gasification increases environmental impacts in  
655 general. Such increases can be mitigated by the self-sufficient energy design, although the self-  
656 sufficient design is no longer preferable when the biorefinery has access to solar and wind. The  
657 environmental impacts are dominated by electricity consumption and CC process. Hydrogen  
658 production via BECCS is a promising option in the near term in terms of economics and the co-  
659 benefits of decarbonizing and reducing environmental impacts in categories such as human health  
660 impacts (including carcinogenics, none-carcinogenics, and respiratory effects), comparing to  
661 fossil-based and electrolysis routes for hydrogen production. The complex trade-offs in technical,  
662 economic, and environmental aspects highlight that the deployment of this BECCS approach  
663 requires endeavors from multi-players from analytics, sustainable biomass supply, chemistry and  
664 engineering, business, and policies.

665 This study has several limitations. CO<sub>2</sub> transportation and storage are not included in this study,  
666 but the LCA and TEA results can lay the foundation for future research comparing BECCS with  
667 other CO<sub>2</sub> removal technologies. Another limitation is the exclusion of hydrogen transportation  
668 that needs to be determined based on hydrogen end use. Risks associated with the handling, storage  
669 and transportation of both H<sub>2</sub> and CO<sub>2</sub> (e.g., hydrogen safety and gas transportation infrastructure  
670 issues) should be considered in future research. It should also be noted that other carbon capture  
671 technologies exist and can be used in combination with gasification (e.g., chemical looping).  
672 Although this study only includes MEA given its high technology maturity, the integrated  
673 modeling approaches presented in this work can be applied to gasification coupled with other  
674 carbon capture technologies. Besides, this study uses process-based TEA and attributional LCA,  
675 therefore economic constraints related to market supply and demand, as well as competing uses of  
676 these biomass are not considered. Future research can include resource constraints and market  
677 effects using ecological-economic models and consequential LCA. Moreover, this study focuses  
678 on the Pacific Northwest, BECCS systems built in other regions may have different environmental  
679 and economic performance due to differences in biomass characteristics and background  
680 processes. The geographic variations should be considered when applying the conclusions of this  
681 study to other regions. Similar to previous LCAs of forest residue utilization (Lan et al., 2022,  
682 2021b), the impacts of forest residue removal on forest ecosystems, e.g., biodiversity, forest fires,  
683 and soil carbon, are not included due to the lack of quantitative data. Recent studies (Dale et al.,  
684 2017; James et al., 2021; Kenderdine et al., 2022) show potential benefits/risks of removing  
685 excessive forest residues, which should be explored in future LCA and TEA.

686

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#### 688 **Declaration of Competing Interest**

689 The authors declare no competing financial interests to affect the work reported in this paper.

690

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702 **Supplementary materials**

703 Supplementary material associated with this article is available.

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