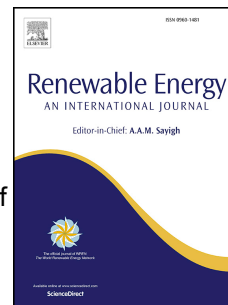# Journal Pre-proof

Comparative techno-economic modelling of large-scale thermochemical biohydrogen production technologies to fuel public buses: A case study of West Midlands region of England

Danielle J. Nouwe Edou, Jude A. Onwudili

Please cite this article as: Nouwe Edou DJ, Onwudili JA, Comparative techno-economic modelling of large-scale thermochemical biohydrogen production technologies to fuel public buses: A case study of West Midlands region of England, *Renewable Energy* (2022), doi: https://doi.org/10.1016/j.renene.2022.02.074.

This is a PDF file of an article that has undergone enhancements after acceptance, such as the addition of a cover page and metadata, and formatting for readability, but it is not yet the definitive version of record. This version will undergo additional copyediting, typesetting and review before it is published in its final form, but we are providing this version to give early visibility of the article. Please note that, during the production process, errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

Danielle J. Nouwe Edou: contributed to the process simulation, process modelling, generation of economic analysis data and drafting of report and editing of the manuscript.

Jude A. Onwudili: contribution included project conceptualisation, methodology, synthesis and supervision of process design and economic analysis; project administration, and writing of the paper (draft, review and final editing).

1

Comparative techno-economic modelling of large-scale thermochemical biohydrogen production technologies to fuel public buses: A case study of West Midlands region of England

Danielle J. Nouwe Edou[1], Jude A. Onwudili* [1,2]

[1]Department of Chemical Engineering and Applied Chemistry, College of Engineering and Physical Sciences, Aston University, Birmingham, B4 7ET, United Kingdom

[2]Energy and Bioproducts Research Institute, College of Engineering and Physical Sciences, Aston University, Birmingham, B4 7ET, United Kingdom

Abstract

This work presents techno-economic modelling of four thermochemical technologies that could produce over 22,000 tonnes/year of hydrogen from biomass for >2000 public transport buses. These included fluidised bed (FB) gasification, fast pyrolysis-FB gasification, fast pyrolysis-steam reforming, and steam reforming of biogas from anaerobic digestion (AD). Each plant was modelled on ASPEN plus with and without carbon capture and storage (CCS), and their process flow diagrams, mass and energy balances used for economic modelling. Payback periods ranged from 5.10 to 7.18 years. For operations with CCS, in which the captured CO_2 was sold, FB gasification gave the lowest minimum hydrogen selling price of $3.40/kg. This was followed by AD-biogas reforming ($4.20/kg), while pyrolysis-gasification and pyrolysis-reforming giving $4.83/kg and $7.30/kg, respectively. Hydrogen selling prices were sensitive to raw material costs and internal rates of return, while revenue from selling CO_2 was very important to make biohydrogen production cost competitive. FB gasification and AD-biogas reforming with CCS could deliver hydrogen at less than or around $4/kg when CO_2 was sold at above $75/tonne. This study showed that four thermochemical technologies could contribute

27   to cheap biohydrogen production to extend the current use of electrolytic hydrogen-fuelled

28   buses in Birmingham to the wider West Midlands region.

29
30   **Keywords:** green biohydrogen fuel, FB gasification, pyrolysis-reforming, pyrolysis-gasification,

31   AD-biogas reforming, techno-economic modelling

32
33
34   *Corresponding Author: j.onwudili@aston.ac.uk
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65
66
67
68

69 **Nomenclature**

70 AD: Anaerobic Digestion

71 ASPEN: Advanced Simulator for Process Engineering (Computer Software)

72 BFB: Bubbling Fluidised Bed

73 AD-BSR: Anaerobic Digestion-Biogas Steam Reforming

74 CCS: Carbon Capture and Sequestration/Storage

75 CFB: Circulating Fluidised Bed

76 FB: Fluidised Bed

77 FCI: Fixed Capital Investment

78 H_2: Hydrogen

79 HT: High Temperature

80 IRR: Internal Rate of Return

81 LT: Low Temperature

82 Mtoe: Millions of Tonnes of Oil Equivalent

83 NCF: Net Cash Flow

84 NPV: Net Present Value

85 PFD: Process Flow Diagram

86 PSA: Pressure Swing Adsorption

87 RME: Rapeseed Methyl Ester

88 SMR: Steam Methane Reforming

89 WGS: Water Gas Shift

90

91

1. Introduction

Increasing global population and improving living standards are major contributors to the exponential increase in global energy demand of approximately 14,000 Mtoe [1, 2] in 2019. Climate change and poor air quality are two of the most severe deleterious effects of our overwhelming dependence and utilisation of fossil fuels. Deployment of commercial-scale sustainable and renewable alternative fuels will help to meet the global 2050 decarbonisation and Net Zero targets [3] and tackle poor air quality in urban centres. Some urban areas, including Birmingham City, in the United Kingdom (UK) are using the Clean Air Zone scheme to discourage the use of old highly polluting vehicles within classified zones [4].

With no harmful emissions at the point of use and a potentially large availability of resources, hydrogen is now regarded as a promising ecologically clean energy carrier capable of ameliorating the long-term energy trilemma challenge. According to the EC Clean Vehicles Directive (Directive (EU) 2019/1161 by 2025 and 2030, EU countries must procure clean buses, including hydrogen-fuelled buses to meet binding emission targets) [5]. This has prompted several public and private initiatives to accelerate the pace of penetration of hydrogen-fuelled buses in Europe's cities and regions. These include the H2BusEurope, JIVE, JIVE2, MERHLIN and 3MOTION schemes and projects that involve deploying about 1322 hydrogen-fuelled and fuel cell electric buses (FCEBs) across 22 cities across Europe by the early 2020s [6, 7].

Following Brexit, the UK is implementing subsisting or similar regulations that are well aligned to the EU Directives using schemes such as the FutureGrid and Project Union projects [8]. Hydrogen-fuelled public bus transport is being advocated in the UK, especially for large cities, including in Birmingham City [9]. By extension, these efforts should lead to replacing all public buses within the wider West Midlands region that are currently running on fossil fuels

115 (mainly diesel). With such initiatives, the demand for hydrogen energy is expected to increase

116 dramatically, with an estimated global consumption anticipated to reach 300 million tonnes by

117 2050 [10,11].

118   Thus, producing hydrogen from an array of renewable sources is currently a hot topic.

119 Depending on the feedstock and/or source of energy input for its production, the final hydrogen

120 product can be colour-coded using a so-called hydrogen colour spectrum from black to white

121 [12]. Generally, the commercial technologies for producing dark-coloured hydrogen (grey and

122 black) include steam methane reforming (SMR) [13], partial oxidation (POX) of methane and

123 other hydrocarbons [13] and coal gasification; all without carbon capture [14]. However, with a

124 tail-pipe carbon capture and storage (CCS) and carbon capture, usage, and storage (CCUS),

125 the colour of hydrogen product from these processes may move to lighter colour shades e.g.,

126 blue hydrogen from SMR with CCS. The ultimate green hydrogen comes from electrolysis of

127 water powered with zero carbon emissions e.g., using wind or solar energy [15].

128   Biomass is a good feedstock for hydrogen production via different processing routes. Being

129 renewable, biomass can become a source of green hydrogen, if the biomass feedstock is

130 sustainably sourced and if the hydrogen production process is combined with CCS and CCUS

131 [16]. Promising technologies for hydrogen production from biomass include conventional

132 gasification [17,18], pyrolysis-gasification [19,20], pyrolysis-reforming [21,22], anaerobic

133 digestion (AD) biogas reforming [23,24] and hydrothermal gasification [25–27]. Recent detailed

134 reviews of these various technologies can be found in literature [18,28]. Within each technology

135 space, different configurations and variants exist; for example, conventional gasification alone

136 has several variants mainly depending on the type of gasifier and the gasifying agent [29]. For

137 instance, in terms of gasifier type, conventional gasification can be classified into downdraft

138 fixed bed, updraft fixed bed, entrained flow, circulating fluidised bed, bubbling fluidised bed and

139 plasma gasification [29,30]. However, the common feature of these technologies is the

140 application of heat and various chemistries to convert biomass feedstocks into final products

141 composed mainly of hydrogen and carbon dioxide, along with small amounts methane, carbon

142 monoxide and even smaller amounts of other hydrocarbon gases [30,31].

143   In this present study, four different technologies for producing hydrogen from biomass have

144 been selected for techno-economic evaluation based on established or growing commercial

145 interests in their large-scale deployment [32–35]. Therefore, these four technologies arguably

146 have the potential to meet the fuel requirement to power the current over 2000 buses being

147 used for public transport in the West Midlands, UK [36,37]. Literature shows that previous

148 researchers have modelled the techno-economic assessment of various hydrogen production

149 technologies [38,39] however, few have combined all the viable biohydrogen production

150 technologies within the context of a realistic public transport scenario. A combination of ASPEN

151 modelling and techno-economic factors have been used to generate relevant process data in

152 this study. It is the aim of this novel study to provide essential chemical engineering data for

153 potential technology providers on large-scale biohydrogen production that will extend beyond

154 the West Midlands public transport system towards meeting Net Zero targets.

2. Methodology

2.1. Design basis and biohydrogen requirement

157 In this section, a detailed outline of assumptions made for ASPEN simulation of the four

158 biohydrogen production technologies (gasification, pyrolysis-gasification, pyrolysis-reforming

159 and AD-biogas steam reforming), are presented, with and without CCS. The ASPEN Software

160 used was ASPEN Plus V11 available at Aston University, Birmingham (UK). Appropriate

161 feedstocks were selected for the different technologies based on literature and their availability

162 to meet the demand. Initial chemical process synthesis was carried out, followed by

163 computation of process flowsheets to obtain mass and energy balances using ASPEN.

164 Furthermore, the data obtained from the ASPEN simulation, along with conventional factors

165 were used to evaluate the economic feasibility of using biohydrogen to meet West Midlands'

166 bus clean transport fuel demand. The impact of implementing CCS on the economic

167 performance of each technology was also evaluated.

168 At present, the West Midlands currently operates over 2,000 buses, travelling a total distance

169 of 515,000 km per day and consuming 230,000 litres of diesel [37,40]. Direct communication

170 with Transport for West Midlands and Van Hool (one of the largest bus operators in Birmingham

171 City), gave the number of public buses as 2000 and 2300, respectively [36,37]. Additionally, on

172 average, 12-m buses consume 9 kg of hydrogen per 100 km [36]. Using these data, the yearly

173 capacity of a hydrogen production plant operating for 330 days a year to produce fuel for 2000

174 buses was calculated as shown in Table 1. In addition, to account for the uncertainty in the

175 actual number of buses and also accommodate potential future expansion, hydrogen

176 production capacity was increased by a factor of 1.25, giving an adjusted hydrogen production

177 rate of nearly 2782 kg/h.

178 Table 1: Case study data used for hydrogen production for 2000 buses [36,39, 40]

Production Details	Value	Units
Average hydrogen consumption by buses	0.09	kg/km
Distance travelled by Bus	257.5	km/day
No. of Days the Bus is Driven	330	days/year
No. of Buses in West Midlands, England	2300	-
Yearly hydrogen requirement	17589.83	tonnes/year
Adjusted yearly hydrogen requirement	22031.78	tonnes/day
Yearly hydrogen plant operating hours	7920	hours/year
Adjusted hydrogen production capacity	2781.79	kg/h
Density of hydrogen at SATP	0.0813	kg/m³
Volumetric production capacity	34216.36	m³/h

179

180 **2.2. Justification of selected process technologies**

181 In this present study, thermochemical processing technologies for biohydrogen production,

182 have been mostly considered due to their technical maturity, potential for large-scale

183 application, high throughput capacity, fast reaction rates and robustness to meet the hydrogen

184 requirement of 22,031.78 tonnes/year. All the processes considered are either at commercial

185 scale or have the potential to reach commercial scale in the next few years. These include FB

186 biomass gasification using circulating fluidised bed (CFB) reactor, combination of CFB pyrolysis

187 and gasification (pyrolysis-gasification), combination of CFB pyrolysis and steam reforming

188 (pyrolysis-reforming) and steam reforming of anaerobic digestion biogas (AD-biogas

189 reforming).

190 Circulating fluidised beds are the most commonly used reactors in commercial gasification

191 plants due to advantages of attaining isothermal conditions devoid of hotspots, excellent mass

192 transfer rates and fast reaction rates [41]. Tar formation is a key challenge, but the use of

193 dolomite and other catalysts can be used to induce tar cracking [42]. Furthermore, the process

194 produces a syngas that mainly consists of hydrogen and carbon monoxide, with hydrogen

195 making up around 40% of the volumetric fraction [39]. The relatively high CO composition of

196 the product gas also gives opportunity for more hydrogen production via water-gas shift (WGS)

197 reactions.

198 During pyrolysis-gasification biomass feedstock first undergoes fast pyrolysis upstream at 500

199 °C using a circulating bed pyrolyser, to produce up to 88 wt.% of vapour-phase products

200 comprising of condensable and non-condensable compounds [43]. The vapour-phase products

201 may be sent directly into a steam gasifier, to produce syngas containing hydrogen and for

202 further downstream hydrogen production via a WGS reactor, condenser, scrubber, and a PSA

203 unit [44]. Otherwise, the vapour-phase product may be separated by quenching and only the

204 liquid product (about 75 wt%) [43] re-vapourised and fed into the steam gasifier. Pyrolysis-

205 reforming is another process route that upgrades the pyrolysis products using catalytic steam

206 reforming. In most cases, cheap and readily available catalysts, such as Ni/Al_2O_3, are used to

207 increase gas yields and thus, a higher production of hydrogen in the syngas from the pyrolysis

208 vapours, largely based on SMR technology. Finally, anaerobic digestion (AD) is used to

209 produce biogas, constituting of about 60% CH_4 and 40% CO_2 [45]. Once biogas is obtained,

210 the CO_2 is removed and the methane-rich biogas is fed into a steam reformer for steam

211 methane reforming (SMR), which is the current most widely used hydrogen production

212 technology, generating around 50% of global hydrogen demand [46].

213 *Delimitations:* The evaluation of each model started with the feeding of solid biomass or biogas

214 of the stated specification into the first thermal conversion equipment, respectively. Each model

215 ended with: (1) the output of the required amount of hydrogen calculated from the design basis

216 (2) where applicable, the combustion of char product to provide fractional process heats for the

217 respective biomass conversion equipment (gasifier and pyrolyzer).

218

219 **2.3. Feedstock selection and justification**

220 Gasification, pyrolysis-gasification, and pyrolysis-reforming use biomass feedstocks in the form

221 of wood chips and wood pellets, of which wood pellets are the most used. The UK produced

222 only 0.3 million tonnes of wood pellets in 2020 and imported 9.1 million tonnes during the same

223 period [47]. Therefore, the process plants based on direct thermochemical processing of

224 biomass for biohydrogen production would rely on biomass imports. In contrast, AD could rely

225 on abundant food and agricultural wastes generated within the UK, which generates around

226 13.1 million tonnes of food waste annually [48]. Therefore, UK has a relatively vast resource to

227 attain resource sufficiency for AD and in this study maize silage has been selected for the AD-

228 biogas reforming plant.

229

230 **2.4 Biomass feedstock characteristics**

231 The average compositions of wood pellets (Supplementary Information A Table SI1) were

232 obtained from literature [39,49] and used for the simulation of feedstock for hypothetical

233 gasification, pyrolysis-gasification, and pyrolysis-reforming plants. Also, the ultimate

234 compositions [50] of the maize silage used as feedstock for the AD-biogas process

235 (Supplementary Information A Table SI1) but this was not used for simulation due to limitation

236 of ASPEN Plus to simulate AD process. However, for simulation, the biogas produced from

237 corn silage was used, since the biomass would be fed into an AD plant, which could not be

238 simulated on ASPEN. Both feedstocks were assumed to have negligible sulphur contents

239 based on literature data of around 0.1%.

240

241 2.5 Process description and syntheses

242 The simulation of each process flow diagram has been presented in Figures 1 - 4, with

243 each having the additional optional CCS stage. Early trials indicated that the three

244 thermochemical processes and the SMR part of the AD-biogas reforming process could be

245 simulated on ASPEN Plus. Therefore, detailed process syntheses were carried out for the

246 construction of process flowsheets with respect to the pieces of equipment to achieve the

247 technical requirements for each process. The simulation of the AD process itself was

248 considered beyond the scope of this present study, but details of biomass conversion and

249 biogas yields for typical AD plants were obtained from literature [39] and used as appropriate.

250

251   2.5.1 FB gasification process

252   Physical properties for the non-conventional, conventional and CISOLID substances were

253   calculated using IDEAL property methods. Furthermore, the enthalpy and density of the non-

254   conventional components, (biomass, char, and ash) were all identified by the HCOALGEN and

255   DCOALIGT models. The process flow diagram for the gasification process is presented in

256   Figure 1. In the figure, an RYield block is used to portray the gasifier, operating at 850°C, where

257   biomass reacts with steam at a steam to carbon molar ratio of 0.4 [39]. Thereafter, an SSplit

258   block representing the cyclone separates the gaseous products (DEC-GAS) being transferred

259   to the heat exchanger (HE), from the solid residues (DEC-CHAR) entering the RYield

260   decomposition (DECOMP) unit. The DECOMP promotes the breakdown of biochar into C, N

261   and ash which passes into an RStoic block where they react with air and the PSA recycle

262   stream to undergo combustion to meet gasification energy demands. Therefore, at steady state

263   the gasifier is assumed to be energetically self-sustaining. The oxygen to carbon molar ratio for

264   the combustor is set at 0.3 [39] and maintaining the oxygen to nitrogen ratio of 21:79 in the feed

265   air. A high temperature water-gas shift (HWGS) reactor was represented by an REquil block,

266   while an Ryield block represented the low temperature water-gas shift (LWGS) reactor, and all

267   heat exchangers by the heater block (Dryer, HE, HE1). Also, the RME scrubber was replicated

268   by the Sep2 block. After separation, the hydrogen and carbon dioxide were compressed in the

269   MCompr blocks (C-1, C-2). Heat transfer between the gasification units was also included within

270   the simulation by heat streams (Q-SUPPLY, Q-SPARED).

271
272
273
274
275
276
277
278

279
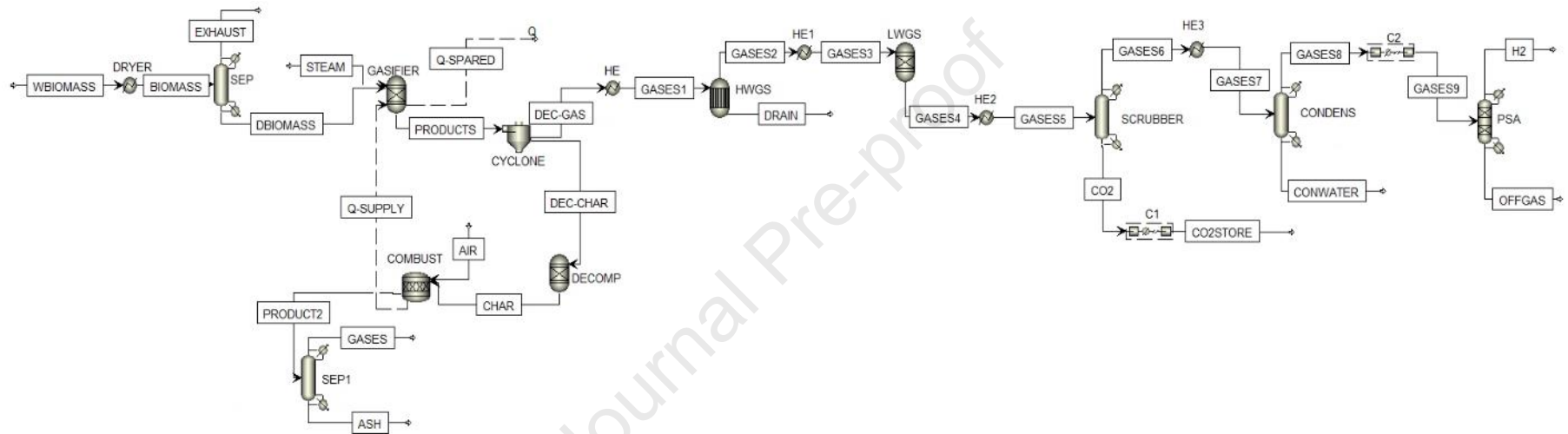
Figure 1. An ASPEN Plus simulation depicting the Process Flow Diagram of Hydrogen Production from Biomass Gasification with Carbon Capture and Sequestration.

WBIOMASS=Wet Biomass; SEP= Separator; DBIOMASS= Dry Biomass; Q = Heat; DEC-GAS= Decomposed Gas; DEC-CHAR= Decomposed Char; DECOMP= Decomposition Reactor; COMBUST= Combustion Reactor; HWGS= High Temperature Water Gas Shift Reactor; LWGS= Low Temperature Water Gas Shift Reactor; CONWATER= Condensed Water; C-1= C-2= Compressor

286   2.5.2 Pyrolysis-gasification process

287   The physical properties, property methods and models were identical to that of gasification.

288   Figure 2 presents the process flow diagram for the pyrolysis-gasification process. The biomass

289   was dried in a heater block (Dryer) at 110 °C to a moisture content of 7%. This was followed by

290   an RYield fluidised bed fast pyrolysis reactor operating at 500 °C [49] under inert conditions,

291   forming char, bio-oil, and gases. The products were assumed to be separated and only the bio-

292   oil used for the gasification step; to eliminate the large volume fluidising gas (nitrogen) and

293   process water. The normalised yields of the syngas, oil, char, ash and water products based

294   on the work of Ringer et al. [49] were used for this stage. Additionally, inlet and outlet heat

295   streams, Q-SUPPLY and Q-SPARED, respectively, were inserted. The solid char was isolated

296   using an SSplit cyclone unit and transferred to an RYield decomposition and RStoic combustor

297   reactor at 600°C in series. Air was fed into the combustor at an air to carbon molar ratio of 1.15

298   [49]. A heater block at 850°C was used to heat up the captured bio-oil, prior to it being fed into

299   the RYield fluidised bed gasification reactor. The steam reacted with bio-oil in the gasifier at a

300   steam to carbon molar ratio of 2.75 [49]. The rest of the process, after the gasification stage,

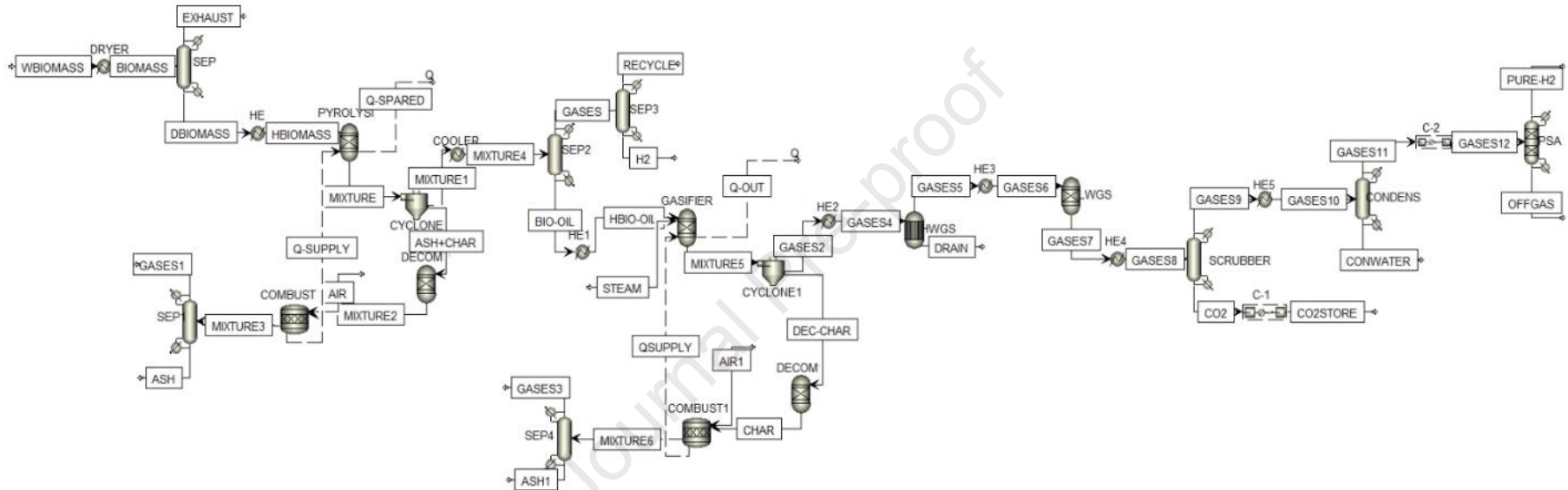301   was assumed to be the same as described in biomass gasification.

302

303
304
305
306
307
308

309
310 Figure 2. An ASPEN Plus simulation highlighting the Process Flow Diagram of Hydrogen Production from Pyrolysis-Gasification
311 with Carbon Capture and Sequestration.

312 WBIOMASS=Wet Biomass; SEP= Separator; DBIOMASS= Dry Biomass; HBIOMASS= Hot Biomass; PYROLYSI= Pyrolysis
313 Reactor; Q = Heat Supply; DECOMP= DECOM1= Decomposition Reactor; COMBUST= Combustion Reactor; HBIO-OIL= Hot
314 Bio-oil; DEC-CHAR= Decomposed Char; HWGS= High Temperature Water Gas Shift Reactor; LWGS= Low Temperature
315 Water Gas Shift Reactor; CONWATER= Condensed Water; CO22= CO2, C-1= C-2= Compressor

316
317

318 2.5.3 Pyrolysis-reforming process

319 The starting point of this process was identical to the pyrolysis-gasification process, beginning

320 with a fluidised based fast pyrolysis process as shown in Figure 3. Hence, a similar set of

321 equipment was needed for this stage. After the pyrolysis stage, the reforming of the re-heated

322 bio-oil would take place in an RGibbs catalytic steam reformer operating at 900 °C. The bio-oil

323 would react with steam, at a steam to carbon molar ratio of 2.75 [49]. Gaseous products leaving

324 the reformer would be cooled by heater block (HE2) to 450 °C, before entering the REquil

325 HWGS reactor at 1.1 bar, followed by an LWGS at 250 °C. A Sep2 (CONDENS) unit would be

326 used to remove water and other condensable components, leaving a gas stream being fed into

327 optional Sep2 (scrubber) to remove CO_2, with the remaining gases compressed to 15 bar and

328 25 °C by an MCompr block (C-1). The specification for the HWGS was the same as that for

329 both biomass gasification and pyrolysis-gasification. The compressed gases would be fed into

330 another Sep2 (PSA) block, where an assumed 99.9% of hydrogen gas would be obtained.

331

332

333

334
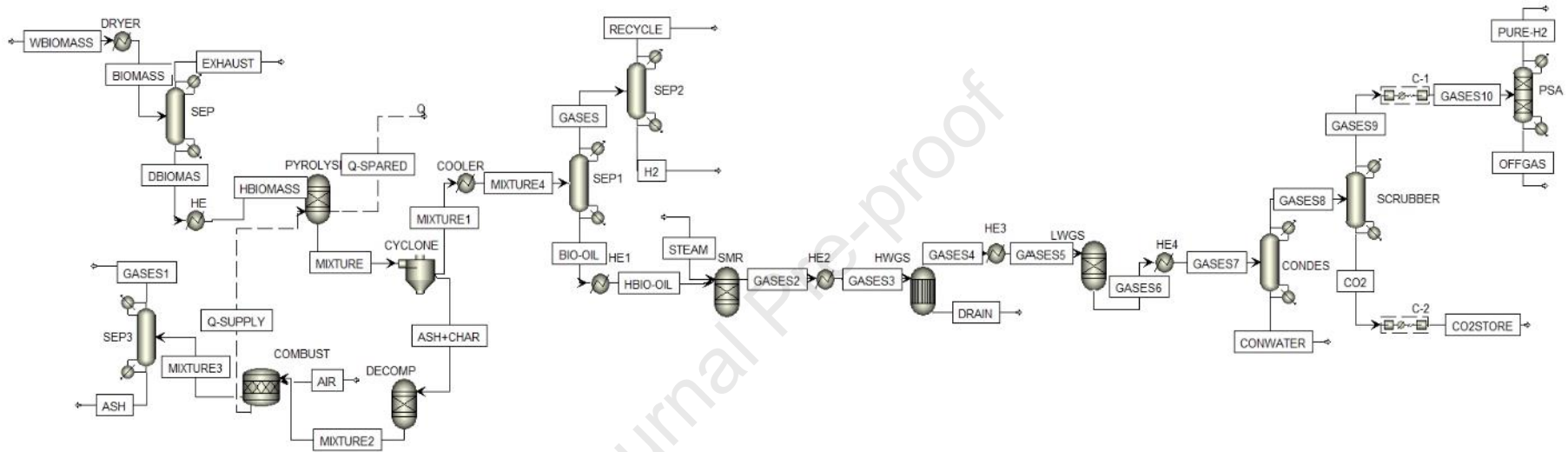
335
336
Figure 3. An ASPEN Plus simulation picturing the Process Flow Diagram of Hydrogen Production from Pyrolysis-Reforming with Carbon Capture and Sequestration.

WBIOMASS=Wet Biomass; SEP= Separator; DBIOMASS= Dry Biomass; HBIOMASS= Hot Biomass; PYROLYSI= Pyrolysis Reactor; Q-SUPPLY= Heat Supply; Q = Heat Spared; DECOMP = Decomposition Reactor; COMBUST= Combustion Reactor; HBIO-OIL= Hot Bio-oil; SMR= Steam Methane Reforming Reactor; HWGS= High Temperature Water Gas Shift Reactor; LWGS= Low Temperature Water Gas Shift Reactor; CONWATER= Condensed Water; C-1= C-2 = Compressor

343 2.5.4. AD-Biogas steam reforming process

344 As stated at the beginning of this section, the AD-biogas steam reforming process simulation

345 would start with the biogas, as ASPEN could not be used to replicate the enzymes used in AD.

346 The BSR process flow diagram is shown in Figure 4. Therefore, the starting point for simulation

347 was the compression of biogas in an MCompr block (C-1). Yields of the products of anaerobic

348 digestion found in literature [45, 50] were normalised before being entered into ASPEN

349 (Supplementary Information A Table SI2).

350 It was assumed that biogas only consists of CH_4 and CO_2 so only the normalised values for

351 these two components were used in the simulation (Supplementary Information A Table SI2).

352 A Sep2 (scrubber) was used to isolate CO_2 whilst the remaining CH_4 was heated by a heater

353 block (HE) and transferred to the RGibbs (SMR reactor) block at an operating temperature of

354 900 °C. The reaction of methane with steam would occur at a steam to carbon molar ratio of 3

355 [39]. Gases leaving RGibbs were cooled to 350 °C by a heater block (HE1) and again, following

356 the same steps detailed under FB gasification. The Redlich-Kwong-Soave cubic equation with

357 Boston-Mathias alpha function (RKS-BM) property method was used, with the density and

358 enthalpy values calculated using the same models as the previous processes.
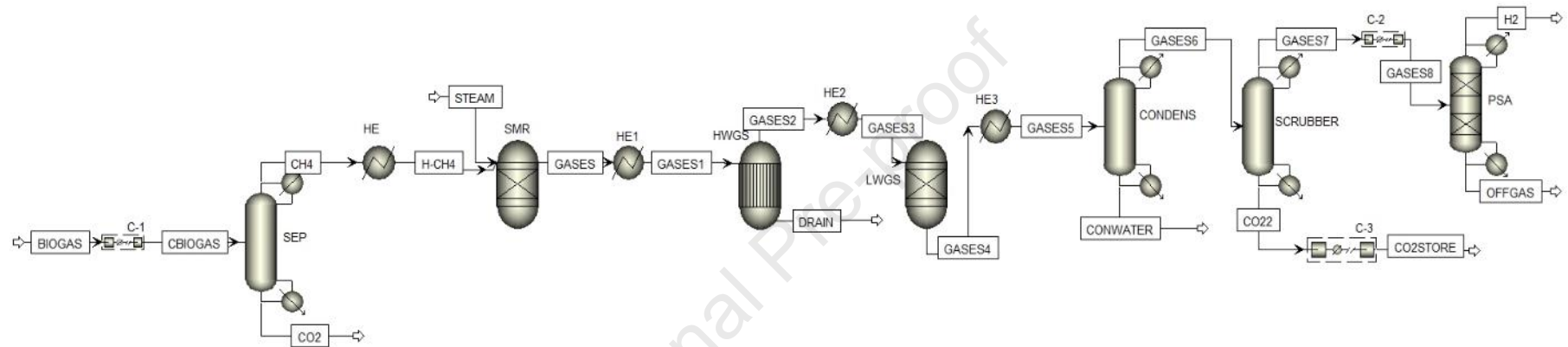
359

360

361
362
363
364
365

366
367 Figure 4. An ASPEN Plus simulation illustrating the Process Flow Diagram of Hydrogen Production from Biogas Steam
368 Reforming with Carbon Capture and Sequestration.

369 CBIOGAS= Compressed Biogas; HE= Heat Exchanger; H-BIOGAS: Hot Biogas; SMR= Steam Methane Reforming Reactor;
370 HWGS= High Temperature Water Gas Shift Reactor; LWGS= Low Temperature Water Gas Shift Reactor; CONDENS=
371 Condenser; CONWATER= Condensed Water; C-1= C-2= C-3= Compressor, PSA= Pressure Swing Adsorption

372
373
374

375

376 For all four processes, an optional unit of an MCompr block (C-2) at 55 bar and 25 °C used

377 to compress and capture CO_2 was included. The flowsheets for processes without carbon

378 capture and storage (CCS) can be found in the Supplementary Information B.

379

380 2.6. Economic analyses

381 This section summarises the description of the approach taken for the techno-economic

382 evaluation of each process route. The following assumptions were used for cost estimation

383 for the base case scenario:

384 - Depreciation was charged over 10 years [51].

385 - Income tax rate was estimated to be 30% [39,51].

386 - Ideal breakeven point was assumed to be 40% [52].

387 - Fluctuations in costs due to general inflation or processing demands were negligible

388   when calculating the Net Present Value [52].

389 - Baseline case discount rate or required return was assumed to be 15% [53].

390 - Where applicable, CO_2 selling price ranged from $0 to $150 per tonne [54]

391 - Wood pellets price was taken as $150 per tonne and maize silage as $45 per tonne;

392   both on 'as received' basis.

393

394 2.6.1. Capital cost

395 All purchased equipment costs were identified using a combination of the ASPEN Process

396 Economic Analyser software and techno-economic studies from literature [52,53]. A detailed

397 factorial method was used to estimate the capital cost of the biomass gasification using US

398 Gulf Coast Basis [52]. Additionally, location factors for USA and the UK were applied to find

399 the realistic capital cost of the same plant in the UK. For instance, published price data from

400   Chemical Engineering Economics [55] were used for fluidized/packed bed reactors to

401   determine cost of equipment. To obtain prices for 2020 (the start of this study), the Oil and

402   Gas Field Machinery, and Equipment Manufacturing Cost Index [56], published in 1989 was

403   used according to Equation 1.

404      $C = C_i \times \dfrac{Index_{2020}}{Index_{1989}} \times f$ (1)

405   Where, C = present cost of equipment, C_i = historical cost of equipment, f = factor accounting for

406   construction material type and insulation (f = 1.30 for stainless steel).

407

408   2.6.2. Operating costs

409   Operating costs in process plants are typically made up of ongoing costs including raw

410   materials costs, maintenance costs, utility costs and labour costs. Apart from feedstock

411   (biomass) costs, the other operating costs have been calculated using typical Lang factors

412   [52] (Supplementary Information A Table SI3)

413

414   2.6.3. Profitability parameters

415   The key economic parameters were calculated [52] using the usual formulae (Supplementary

416   Information A Table SI4). These include depreciation, net cash flow (NCF), breakeven point,

417   payback period, net present value (NPV) and minimum hydrogen selling price. In this study,

418   three main scenarios were considered for each of the four selected technologies: (a) plant

419   operations with CCS and selling captured CO_2; (b) operation with CCS but without selling the

420   captured CO_2; and (c) operation without CCS. The breakeven point was expected to occur at

421   below 70% [32] of the operating capacity to still make profit while accommodating fluctuations

422   in operational contingencies and market demands. Finally, for each scenario, the selling price

423   of H_2 was adjusted to obtain a net present value (NPV) of zero to obtain the minimum selling

424 price of hydrogen for comparison with other commercially viable hydrogen production
425 technologies.

3. Results and Discussion

427 3.1 Mass balances

428 In the chemical industry, raw materials account for 80-90% of the total cash cost of production
429 (CCOP) [57] and it is therefore an important factor to consider during economic evaluation of
430 chemical processes. The biomass feedstock requirements based on ASPEN simulation for
431 each technology are presented in Table 2 as part of Blackbox mass balances. The table
432 shows that the process requiring the largest amount of raw material was gasification with a
433 total of 93,784.84 kg/h biomass (wet basis) to produce the required 2,781.79 kg/h of
434 hydrogen. On the other hand, the technology requiring the least biomass was pyrolysis-
435 gasification, with a total initial biomass quantity of 29,013.67 kg/h (wet basis). The biomass
436 requirement for AD of 69,905.30 kg/h (wet basis) was estimated on the basis of the quantity
437 of biogas and typical conversion efficiencies needed to make the required amount of
438 biomethane to produce the stated amount of hydrogen [39].

439 When combined in series with other upstream processes such as pyrolysis, gasification
440 became more attractive as less raw material (a decrease of 69.1%) was required as seen in
441 the case of pyrolysis-gasification. Moreover, combining pyrolysis in series with reforming
442 required similar biomass feedstock as pyrolysis-gasification. The dramatic reduction in
443 feedstock requirement compared to gasification could be attributed to the increased
444 contribution of hydrogen atoms in the reactant steam to eventual hydrogen gas product [25].
445 The carbon cost of biohydrogen production via thermochemical technologies lies in the
446 inevitable co-production of CO_2, which can influence their economic and environmental

447   performance [58]. CO_2 co-production thus has implications for environmental sustainability (if

448   emitted) and economic viability (if sold) on biomass-based hydrogen production technologies.

449   Clearly, since the gasification process consumed the most biomass H/C ratio = 1.5 in Table

450   SI1), it meant that more of the carbon element would be present throughout. Thus, a greater

451   mass flowrate of carbon dioxide was produced, totalling 705,473 tonnes/year. As expected,

452   the CO_2 production rate decreased with a reduction in biomass feedstock requirement, hence

453   the drop to 408,522 tonnes/year and then 316,801 tonnes/year for pyrolysis-gasification and

454   pyrolysis-reforming, respectively. AD-biogas steam reforming gave a much lower direct CO_2

455   production rate per annum of 234,806 tonnes/year due to using mainly methane (H/C ratio =

456   4) for hydrogen production. The large participation of steam in hydrogen production from the

457   reforming-based technologies, meant that they required lower quantities of biomass

458   feedstocks and thus generating lower CO_2 co-product than gasification-based technologies.

459   Char combustion in the three solid biomass thermal conversion technologies, will emit further

460   CO_2 (Supplementary Information A Table SI5). As char combustion would only provide a

461   fraction of the process heat required for each gasifier and reformer (Supplementary

462   Information A Table SI6), additional biomass combustion would be needed provide the

463   balance. Therefore, CO_2 emissions from the additional biomass combustion must be included

464   in life-cycle analysis studies, which is beyond the scope of this present study. The detailed

465   mass balance calculations and enthalpy flows (heat balances) for each technology, with or

466   without CCS, are given in Supplementary Information B.

467   Table 2: Blackbox mass balances for all four hydrogen production technologies

Streams	Stream Name	FB Gasification		Pyrolysis-Gasification		Pyrolysis-Reforming		Biogas Steam Reforming	
		With CCS	Without CCS	With CCS	Without CCS	With CCS	Without CCS	With CCS	Without CCS
Inlet (kg/h)	Wet biomass	93,784.84		29,013.67		30,504.24		(69,905.30)	
	Dry biomass	(71,049.12)*		(19,342.45)*		(20,336.16)*		(24,466.85)*	
	Steam	21,058.06		40,634.09		42,721.65		18,870.01	
	Biogas	-		-		-		18,232.68	
Total		**114,842.90**		**69,647.76**		**73,225.89**		**37,102.69**	
Outlet (kg/h)	Exhaust from biomass dryer	11,367.86	11,367.86	8,994.24	8,994.24	9,456.32	9,456.32	-	-
	Char (+ Ash)	6006.84	6006.84	6,301.29	6,301.29	3,411.87	3,411.87		
	Condensed water	1,909.99	1,909.99	1,904.04	1,904.04	1,035.46	1,035.46	4,561.15	4,561.15
	H$_2$ Product	2,781.79	2,781.79	2,781.79	2,781.79	2,781.79	2,781.79	2,781.79	2,781.79
	Recycle gases	-	-	4,642.91	4,642.91	4,881.43	4,881.43	-	-
	CO$_2$ to Storage	89,074.89	-	40,000.07	-	51,581.09	-	29,647.17	12,681.98
	Off-gases	3,701.54	92,776.43	5,023.43	45,023.50	78.00	51,659.09	112.59	17,077.78
Total		**114,842.90**	**114,842.90**	**69,647.76**	**69,647.76**	**73,225.89**	**73,225.89**	**37,102.69**	**37,102.69**

468   * Data in parenthesis have not been used in the mass balance calculations

469

470

471 3.2. Enthalpy flows (heat balances)

472

473   ASPEN Plus simulation (Supplementary Information A Table SI6) was used to calculate the

474 enthalpy flows (heat balances) for the main biomass conversion equipment for each process

475 as well as heat provided via residual char combustion, except for AD-biogas reforming. Even

476 with char combustion in applicable technologies, all four technologies presented net positive

477 heat balances due to the main initial biomass or biogas conversion reactions all being highly

478 endothermic. Pyrolysis-gasification gave the highest net heat balance of +101,906.19 kW, due

479 to the presence of both the pyrolyzer and gasifier, which require high heat inputs. Char

480 combustion could be used to provide significant amount of heat energy for the main feedstock

481 conversion reactors, except for AD-biogas reforming (Table SI6). Literature has shown that

482 solid biomass thermal conversion technologies can be energetically self-sufficient by

483 combusting some of the biomass and the inevitable char product [17]. From literature, the char

484 combustor efficiency of 98% was assumed, with flue gas temperature of 1200 °C [59]. The heat

485 exchange temperature approach for the gasifier was assumed to be 100 °C. Therefore,

486 assuming the flue gas temperature reduced to 950 °C after heating the gasifier operating at

487 850 °C, the heat balance of the flue gas across the gasifier was calculated to be -19,368.83

488 kW, using Equation 2.

489 $$\dot{Q}_{flue\,gas} = \sum_{i=1}^{n} n_i C_{p,i} \left(T_2 - T_1 \right) \tag{2}$$

490

491 $\dot{Q}_{flue\,gas} = enthalpy\ balance\ of\ flue\ gas$

492 $n_i = molar\ flow\ rate\ of\ flue\ gas\ component\ i$

493 $C_{p,i} = avaerage\ molal\ specific\ heat\ capacity\ of\ flue\ gas\ component\ i$

494 $T_2 = flue\ gas\ temperature\ after\ heating\ gasifier$

495 $T_1 = temperature\ eof\ flue\ gas\ xiting\ combustor$

496   Similar calculations were performed for the pyrolyzer (operating at 600 °C). As shown in

497   Table SI6, the hot flue gas from the combustion of char could be used to offset some of the

498   process heat, thereby reducing the overall process heat requirements. This will also reduce the

499   amount of biomass combusted for process heat around the solid biomass conversion

500   equipment (gasifier and pyrolyzer). In addition, the spent flue gas (still at 950 °C) can be used

501   to pre-heat the combustion air and/or used in a waste heat boiler to raise steam, as part of

502   energy integration. Detailed analysis of the energy efficiencies and influence on hydrogen

503   selling price across each technology would include other forms of energy (mechanical and

504   electrical), which is outside the scope of this present study. The use of typical Lang factors [52]

505   to represent cost contributions from utilities and services (including energy) obviates the need

506   for such complicated analysis in this type of study.

507

508   3.3 Economic modelling results

509   This section presents an economic evaluation of each of the four selected hydrogen production

510   technologies by obtaining the minimum hydrogen selling price for each scenario studied. The

511   key parameters used included the capital costs, operating costs, minimum hydrogen selling

512   price, CO_2 selling price, cumulative net cash flow, net present values, breakeven points, and

513   payback periods. Economic analyses were carried out with and without CCS for each process

514   using a baseline case of 15% discount rate, which is typical of bioenergy projects [53]. In

515   addition, sensitivity analysis results on the influence of raw material costs, discount rates and

516   CO_2 selling price on the minimum selling price of hydrogen.

517   3.3.1 Total production costs

518 Through comparison between the costs provided by the computer software and data obtained

519 in literature [60, 61] a final purchasing cost for a circulating fluidised bed (CFB) reactor was

520 calculated by order of magnitude for gasification, pyrolysis-gasification and pyrolysis-reforming,

521 respectively. An example of evaluated total capital expenditure for each selected hydrogen

522 production technology (with CCS) can be found in the Supplementary Information A (Table

523 SI7). The overall production costs and other economic parameters used for the modelling of

524 the four plants under different scenarios were also calculated (Supplementary Information A

525 Table SI8).

526 Figure 5 shows the total production costs required for each technology to produce the 22,032

527 tonnes per year of hydrogen needed to fuel all 2,000 buses in West Midlands, England. The

528 TPC was broken down into capital and operating costs as shown in Figure 5. As mentioned in

529 Section 2.5.4, the ASPEN simulation of the AD-biogas reforming technology did not include the

530 upstream AD plant, however, the purchased cost of AD was obtained from literature [62] and

531 included in the economic analysis. Even with the addition of this unit, AD-biogas reforming

532 remained the cheapest process with a capital cost of $101.24 million with CCS. In a similar

533 way, FB gasification with fewer processing equipment also gave a lower capital cost than

534 pyrolysis-gasification and pyrolysis-reforming. Unsurprisingly, pyrolysis-gasification with and

535 without CCS were found to be cheaper than pyrolysis-reforming by $54.69 million and $43.03

536 million, respectively.

537
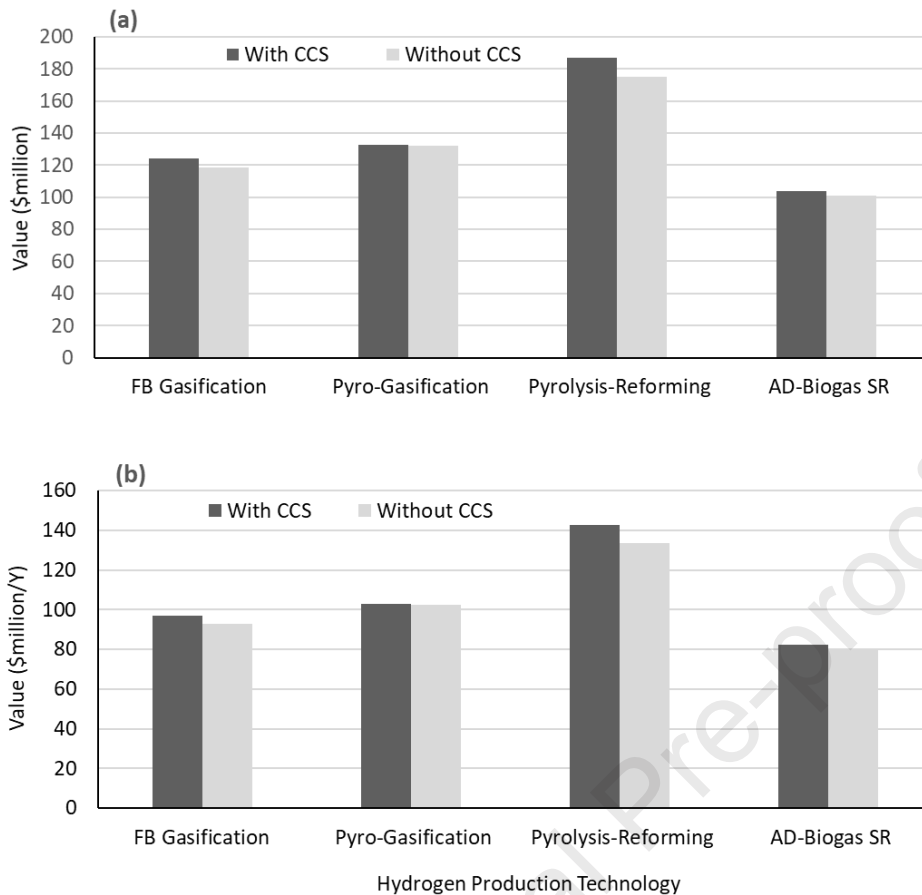
Figure 5. Hydrogen production costs for all four technologies without and with CCS (a) capital

costs; and (b) operating costs

A similar trend was also observed for the operating costs, such that biogas-steam reforming

gave the lowest operating costs, followed by FB gasification. Even though FB gasification

required the largest quantity of biomass feedstock to produce the required amount of hydrogen

(Table 2), it still gave a lower operating cost than pyrolysis-gasification and pyrolysis-reforming,

respectively, indicating that the complexity of the processes contributed more to the operating

costs. Therefore, at the scale required, the two hyphenated thermochemical biomass

conversion technologies (pyrolysis-gasification and pyrolysis-reforming), requiring two large

547 initial reactors each, appear to be highly expensive compared to FB gasification and AD-biogas

548 reforming.

549 Between the two hyphenated thermochemical processes, pyrolysis-reforming was much more

550 expensive than pyrolysis-gasification, with or without CCS. With CCS, the capital cost and

551 operating costs of pyrolysis-reforming were, respectively, 24.6% and 27.6% higher than

552 pyrolysis-gasification. Without CCS, pyrolysis-reforming was also 24.6% and 23.3% higher than

553 pyrolysis-gasification in terms of capital cost and operating costs, respectively. While Table 2

554 shows that both pyrolysis-gasification and pyrolysis-reforming required similar quantities of

555 biomass feedstock and steam, the large difference in total production costs is due to the overall

556 complexity of pyrolysis-reforming technology.

557 3.3.2 Minimum hydrogen selling price

558 The minimum hydrogen selling price was obtained when the NPV of each process became

559 zero, indicating that above that selling price, NPV became positive, which was deemed

560 sufficient to determine the feasibility of each technology at the early stage of engineering

561 projects [52]. For the baseline case, CO_2 selling price was fixed at \$120 per tonne (where

562 applicable), which according to Salkuyeh, Saville and Maclean [63], should be the least selling

563 price of CO_2 to make biohydrogen production from thermochemical technologies cost

564 competitive with SMR (the cheapest hydrogen price in the market).

565 Figure 6 presents the minimum selling prices of hydrogen product for the selected technologies.

566 Results have been evaluated with a consideration to sell or not to sell the captured CO_2 in

567 operations with CCS. Selling CO_2 to obtain low hydrogen selling price may become useful in

568 future with the advancement in in technology for CO_2 utilisation, with potential large-scale CO_2

569 market in the horizon. The CO_2 could be sold to the brewing industry, algae cultivation industry

570   or used for fuel and chemical production; the latter being a subject of increasing research

571   interest. In all cases, breakeven points occurred below 50% operating capacity of each plant

572   (Supplementary Information A Figures SI1 and SI2), indicating huge capacities for the plants to

573   make profit, if the minimum hydrogen selling prices could be competitive. In addition, the

574   payback periods for each scenario for each plant were between 5.10 and 7.18 years

575   (Supplementary Information A Figure SI3) However, lower payback periods were obtained with

576   the implementation of CCS due to increased income from CO_2 sales, which supersedes the

577   additional CCS investment. At the scale of production used in this study, pyrolysis-reforming,

578   and without CCS and with CCS and CO_2 sold, gave lowest payback periods of 5.51 and 5.10

579   years, respectively. Typical payback periods for biomass conversion plants are 2 – 7 years

580   [64,65]. Therefore, all the four technologies (with CCS) selected for this present study were

581   within the typical range of typical payback periods.

582   With CCS and CO_2 being sold as a co-product, all four technologies gave hydrogen selling

583   price of between the lowest value of \$3.40/kg for FB gasification and the highest value of

584   \$7.30/kg for pyrolysis-reforming. Figure 6 however, shows the more present realistic scenarios

585   of CCS without selling CO_2 (e.g., sequestration in old oil formations – with added transport cost

586   not considered in this study) and without CCS at all. For both scenarios, CCS with no CO_2 sold

587   gave slightly higher hydrogen selling price than without CCS, due to the additional total

588   production costs involved in CCS. Capturing CO_2 without selling it still represented the better

589   option to make biohydrogen production greener and comparable to other low-emission

590   hydrogen production routes (e.g., electrolysis of water). Therefore, in this scenario, AD-biogas

591   reforming gave the lowest minimum hydrogen selling price of \$4.75/kg, followed by FB

592   gasification at \$5.50/kg, with pyrolysis-reforming and pyrolysis-gasification giving \$8.10/kg and

593   \$6.1/kg, respectively. Although, Figure 6 shows that minimum hydrogen selling prices were

594 fractionally lower for all four technologies without CCS, this scenario should be deemed as

595 being environmentally unsustainable and therefore not encouraged.

596
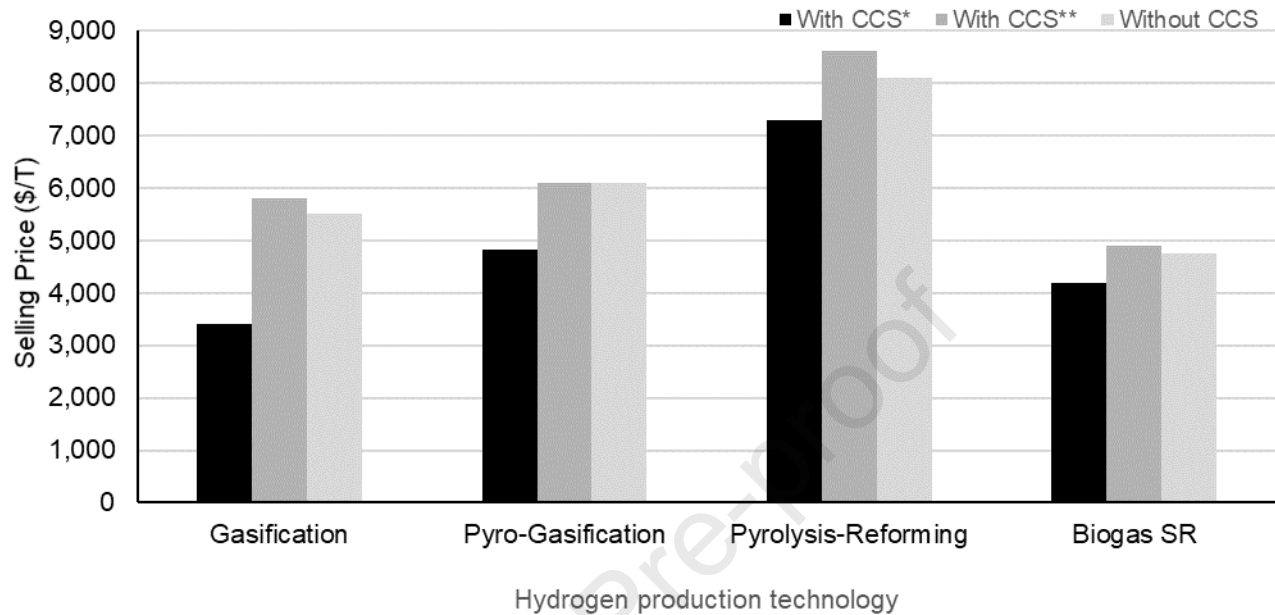
597
598
599 Figure 6. Hydrogen selling price for each production technology without and with CCS
600 (* selling the captured CO_2; ** not selling the captured CO_2)
601
602

603 These results show that at scale, both FB gasification ($3.4/kg) and AD-biogas reforming

604 ($4.20/kg) compared favourably with the current market prices of biohydrogen from

605 manufacturers such as the Canadian company, H_2 V Energies, who sell hydrogen at $2.67/kg

606 [66]. In addition, Salkuyeh, Saville, and MacLean [43] obtained a value of $3.10/kg as the minimum

607 selling price of hydrogen from a fluidized bed gasification process (FB gasification), which is

608 similar to the value obtained from this present study.

609 Although, the minimum selling prices from the four technologies were still higher than the selling

610 price of highly polluting grey hydrogen from SMR at $1.50 [67], significant future investments

611 could help make them more competitive. In comparison, green hydrogen from water

612 electrolysis, currently sells at around $12/kg and future sustainable prices have been predicted

613 to be between $6 and $7 per kg depending on source of electricity [67]. Therefore, this present

614 study shows that thermochemical production of biohydrogen with CCS is potentially a relatively

615 cheap route to green hydrogen using FB gasification and AD-biogas reforming, whereas green

616 biohydrogen from pyrolysis-gasification and pyrolysis-reforming are at close parity to water

617 electrolysis. However, the large quantities of CO_2 generated from these technologies must

618 deliver economic value through sales in order for these biohydrogen routes to be highly

619 successful economically. Hence, there is a wide range of research activities going on to find

620 economic uses of CO_2.

621

622 3.4 Results of sensitivity analyses

623 3.4.1 Influence of raw material costs

624 Figure 7 shows the influence of varying the raw material costs on the minimum hydrogen selling

625 price. The costs of the main raw materials (biomass feedstock, water, steam and catalysts)

626 have been varied by +/-20% of the nominal values obtained from the initial economic modelling

627 of the four hydrogen production technologies. The range have been chosen as realistic

628 scenarios considering that with the limitations on economic production of biomass feedstocks,

629 it is highly unlikely that prices would fluctuate outside this range by so much. The evaluation

630 was based on the three scenarios used in this present study. With raw material costs falling by

631 20%, the minimum hydrogen selling prices also decreased compared to those obtained with

632 the nominal raw materials costs. In the case of CCS with CO_2 sold, hydrogen minimum selling

633 price fell by 7.6% for pyrolysis-reforming, 8.1% for pyrolysis-gasification, 8.2% for AD-biogas

634 reforming and 12% for FB gasification. These changes in hydrogen minimum selling prices

635 reflected the differences in the quantities of biomass required for each technology. Therefore,

636 with FB gasification requiring the most biomass, the effect of changing raw material costs would

637 have the most impact on the minimum hydrogen selling price from this technology. Hence, the

638 minimum hydrogen selling price fell to $2.50/kg for FB gasification, which although still higher

639 than hydrogen from SMR, was nearly 5 times cheaper than the current price of hydrogen from

640 water electrolysis ($12/kg) [68].

641 It would also remain cheaper even at the future green hydrogen price of between $6 and $7

642 per kg, provided CO_2 sales could be guaranteed. When the raw material costs were increased

643 by 20%, a similar trend was observed where the minimum selling price of hydrogen again

644 increased by 12% for FB gasification, 7.9% for pyrolysis-gasification, and 7.6% each for

645 pyrolysis-reforming and AD-biogas reforming. For the other scenarios, a similar trend was

646 observed with hydrogen minimum selling prices decreasing when raw materials costs fell by

647 20% and also increasing when they increased by 20%. These results show that raw material

648 costs can have significant influence on hydrogen selling prices for all four technologies and

649 through all the studies scenarios.

650

651
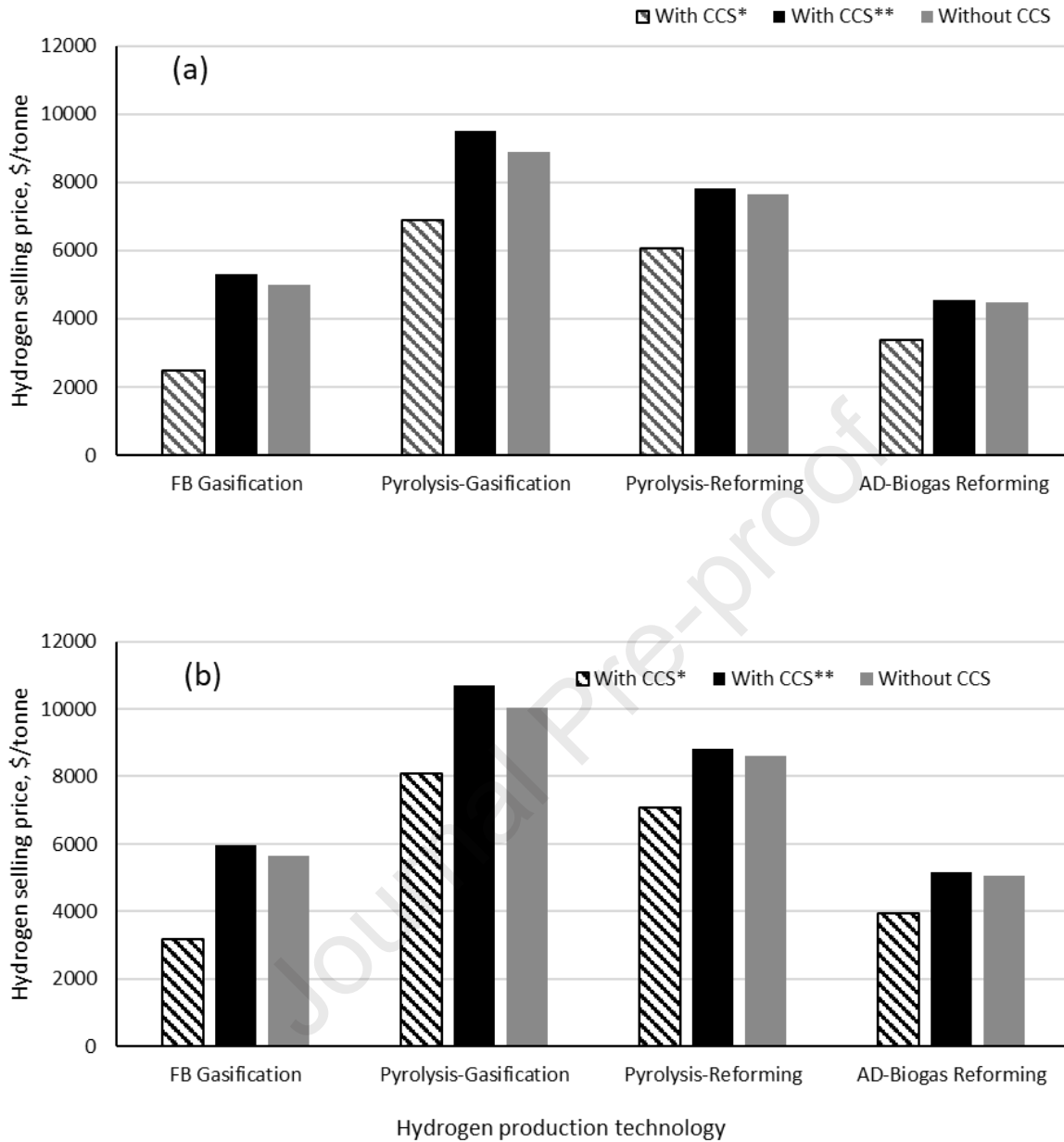
Figure 7. Influence of changes in raw materials cost on the minimum selling price of hydrogen
from the different technologies (a) at -20%; and (b) at +20% of nominal values

654

3.4.2 Influence of discount rate (internal rate of return)

The influence of internal rate of return (IRR) on the minimum hydrogen selling price is presented

in Figure 8. In this study, the internal rate of return was used as discount rate, assumed as the

expected rate of return and then used to find the minimum selling price of hydrogen that gave

659 a net present value of zero for each selected technology and scenarios. The IRR was used at

660 10%, 15% (baseline case) and 20% for this present study. Most bioenergy projects are

661 considered high-risk due to lack of long years of technical experience and IRR values can vary

662 widely, however, for waste-to-energy plants, IRR values of around 10% - 15% are seen as

663 acceptable [53, 69,70].

664 It is clear from Figure 8 that the minimum hydrogen selling prices increased linearly for each

665 technology and each scenario. In all three cases, FB gasification with CCS and CO_2 sold, gave

666 minimum hydrogen selling prices of \$2.52/kg at 10%, \$2.84/kg at 15% (baseline case) and

667 \$3.2/kg at 20% IRR, respectively. Similarly, AD-biogas reforming with CCS and selling the

668 captured CO_2 gave minimum hydrogen selling prices below \$4/kg (\$3.42/kg at 10%, \$3.67/kg

669 at 15% and \$3.95/kg at 20%).

670
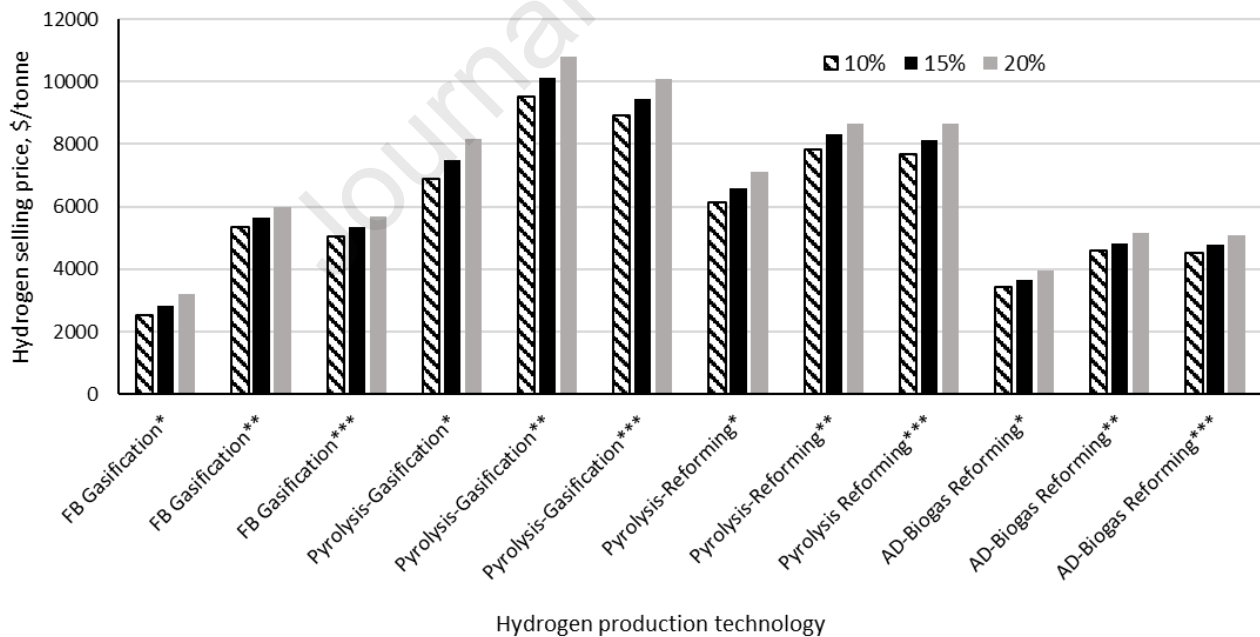
Figure 8. Influence of changes in internal rate of return on the minimum selling price of hydrogen from the different technologies (* with CCS, CO_2 sold; ** with CCS, CO_2 not sold; *** without CCS)

676 Therefore, for both technologies, investors could expect to be cost competitive with attractive

677 IRR of up to 20%. While higher IRRs could be potentially obtained, keeping the minimum selling

678 price of hydrogen around $3/kg or below is important for these technologies to be viable and

679 competitive. For pyrolysis-gasification and pyrolysis-reforming, the minimum hydrogen selling

680 prices remained above $6/kg which seemed high but could be competitive with green hydrogen

681 from water electrolysis.

682

683 3.4.3 Influence of CO_2 selling price

684 As mentioned in Section 3.3, CO_2 production is an investable consequence of hydrogen

685 production from any carbon-based feedstock, including biomass. So far, results have shown

686 that the sale of the CO_2 co-product has significant impact on the minimum hydrogen selling

687 price for each technology. Clearly, with CO_2 captured via CCS and sold of economic utilisation,

688 hydrogen selling price could be lowered to make it more competitive with SMR. Although, CCS

689 would add to the TPC of each technology, the revenue from selling the CO_2 can outweigh this

690 added cost and deliver cheaper biohydrogen. Figure 9 presents the influence of varying the

691 selling price of the captured CO_2 from $0 to $200 per tonne. The results in Figure 9 also

692 demonstrate the effect of the quantity of biomass used and therefore CO_2 expected from each

693 technology. For instance, FB gasification required the largest amount of biomass (93,784.84

694 kg/h in Table 2) and therefore would produce the largest quantity of CO_2 both from the process

695 stream and from char combustion for heat energy. Indeed, Figure 9 shows that the minimum

696 hydrogen selling price fell sharpest with increasing CO_2 selling price for FB gasification and this

697 rate of fall was followed by pyrolysis-gasification, which required the second largest biomass

698 feedstock on dry basis (Table 2).

699
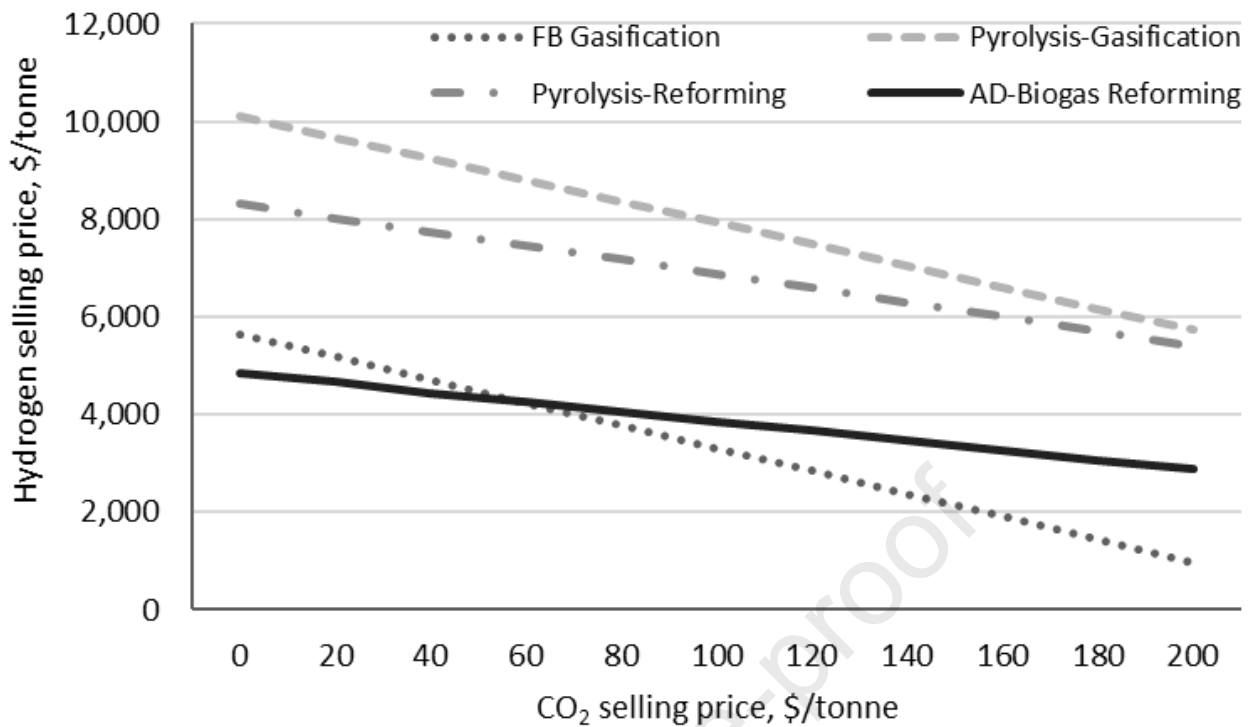
Figure 9. Influence of CO_2 selling price on the minimum hydrogen selling price for the different technologies.

For the reforming technologies, the rate of decreasing minimum hydrogen selling prices were slower due to the large contributions of water in hydrogen production in these processing, which ultimately reduced their biomass feedstock requirements. For FB gasification, the hydrogen selling price fell below \$4/kg when the CO_2 selling price was about \$75/tonne, while for AD-biogas reforming, this could also be achieved around a CO_2 selling price of \$80 - \$85 per tonne. As seen earlier, these two technologies have the better chances of being cost competitive. However, for pyrolysis-gasification and pyrolysis-reforming, the minimum hydrogen price could only break below \$6/kg with a CO_2 selling price of \$160/tonne (pyrolysis-reforming) and \$190/tonne for pyrolysis-gasification. Although these two hyphenated thermochemical technologies appeared to deliver higher minimum hydrogen selling prices, they could compete with hydrogen from water electrolysis. In addition, they both have the advantage of using much

714 lower biomass feedstock compared to FB gasification and may also have the technical

715 advantage of reduced problems with ash and tar at the syngas production stage compared to

716 FB gasification [71].

717

4. Conclusion

719 The slow introduction of hydrogen fuel has resulted from major barriers regarding high

720 feedstock costs, lack of infrastructural development for its production and transport, which

721 appear to outweigh its practical environmental and sustainability benefits. However, it has

722 become apparent that with further supportive policies from governments into the research and

723 development of different thermochemical biohydrogen production technologies, which have the

724 best chances of large-scale capacities, they may attain commercial viability sooner than later.

725 In this study, to identify the most feasible of these technologies process, simulations with and

726 without CCS followed by techno-economic assessments were undertaken. ASPEN Plus was

727 used to simulate the following processes: FB gasification, pyrolysis-gasification, pyrolysis-

728 reforming, and biogas steam reforming.

729 All four technologies are energy-intensive and, apart from AD-biogas reforming, some of the

730 energy requirements can be satisfied from the combustion of the resultant char formed during

731 the initial biomass conversion stage. Despite this fact, FB gasification and AD-biogas reforming

732 could deliver hydrogen at minimum selling prices that are much lower than that currently

733 obtained from water electrolysis. Results show that hydrogen selling prices were sensitive to

734 CO_2 selling price, raw material costs and IRR. However, capturing and selling the CO_2 co-

735 product appeared to be the most important economic factor in diversifying the income streams

736 in order to deliver hydrogen at affordable prices. Although, uncertainties remain in term of the

737 influence of inflation on feedstock prices, equipment costs and labour costs, the scenario

738 presented in this work indicate that biohydrogen production from these technologies, especially

739 FB gasification and AD-biogas reforming, can be delivered affordably and sustainably to power

740 all the public transport buses in West Midlands, England. Technical challenges such as tar

741 formation remain with FB gasification, which could be overcome via pyrolysis-reforming, but

742 this technology require further research and development to become more economically viable.

743

744 **Acknowledgements**

748

749

750

751

752

753

754

755

756

757

758

759

760

761

5. References

[1]   I. Dincer, C. Acar, 2015. Review and Evaluation of Hydrogen Production Methods for Better Sustainability. Int. J. Hydrogen Energy. 40, 11094 – 11111

[2]   The Enerdata Yearbook, 2020. Global Energy Statistical Yearbook 2020. Online: https://yearbook.enerdata.net/total-energy/world-consumption-statistics.html. [Accessed: 01 11 2021]

[3]   IEA, 2021: Net Zero by 2050 – A Road MAP for the Global Energy Sector. Online: https://iea.blob.core.windows.net/assets/20959e2e-7ab8-4f2a-b1c6-4e63387f03a1/NetZeroby2050-ARoadmapfortheGlobalEnergySector_CORR.pdf. [Accessed on 23/06/2021]

[4]   DEFRA, UK., 2020. Guidance: Driving in a Clean Air Zone. Department for Environment, Food & Rural Affairs. Clean Air Zones. Online: https://www.gov.uk/guidance/driving-in-a-clean-air-zone. [Accessed: 14/06/2021]

[5]   European Union, 2019. Directive (EU) 2019/1161 amending Directive 2009/33/EC on the promotion of clean and energy-efficient road transport vehicles

[6]   European Union, 2017. European Clean Bus Deployment Initiative Declaration. Online: https://ec.europa.eu/transport/sites/default/files/2017-05-05-european-clean-bus-deployment-initiative-declaration.pdf [Accessed on 23/06/2021]

[7]   Fuel Cell Electric Buses, 2021. Online at https://www.fuelcellbuses.eu/wiki/policy-framework/eu-clean-bus-deployment-initiative. [Accessed on 22/06/2021]

[8]   National Grid (UK), 2021. The Hydrogen Colour Spectrum. Online: https://www.nationalgrid.com/stories/energy-explained/hydrogen-colour-spectrum. [Accessed on 24/06/2021]

[9]   Birmingham City Council 2020. Hydrogen buses coming to Birmingham next Spring. Online: https://www.birmingham.gov.uk/news/article/727/hydrogen_buses_coming_to_birmingham_next_spring [Accessed 14/10/2020]

[10]   Y. Wang, G. Li, Z. Liu, P. Cui, Z. Zhu, and S. Yang, 2019. Techno-economic analysis of biomass-to-hydrogen process in comparison with coal-to-hydrogen process. Energy. 185, 1063 - 1075.

[11]   IEA, 2015. Energy Technology Perspectives. International Energy Agency (IEA). Paris. Online: https://www.iea.org/reports/energy-technology-perspectives-2015. [Accessed: 12/03/2021]

[12]   J. Dodgshun, 2021. Hydrogen: Clearing up the Colours: From green hydrogen to black — what hydrogen colours mean for the climate Enapter. Online: https://www.enapter.com/hydrogen-clearing-up-the-colours. [Accessed: 14 06 2021]

[13]   M. El-Shafie, S. Kambara, and Y. Hayakawa, 2019. Hydrogen Production Technologies Overview. J. Power and Energy Eng., 7, 107 - 154.

[14]   Office of Energy Efficiency & Renewable Energy, 2020. Hydrogen Production: Coal Gasification; Hydrogen and Fuel Cel Technologies Office. Online:

813   https://www.energy.gov/eere/fuelcells/hydrogen-production-coal-gasification. [Accessed: 05 01
814   2021]
815
816   [15]   Office of Energy Efficiency & Renewable Energy, 2020. Hydrogen Resources. Online:
817   https://www.energy.gov/eere/fuelcells/hydrogen-resources. [Accessed: 14/06/2021]
818
819   [16]   I. Dincer, and C. Zamfirescu, 2012. Sustainable Hydrogen Production Options and the Role of
820   IAHE. Int. J. Hydrogen Energy 37, 16266 - 16286.
821
822   [17]   A.V. Bridgwater, 1995. The Technical and Economic Feasibility of **Biomass Gasification** for
823   Power Generation. Fuel 74 (5), 631-653, 1995
824
825   [18]   V.S. Sikarwar, M. Zhao, P. Clough, J. Yao, X. Zhong, M. Z. Memon, N. Shah, E. J. Anthony and
826   P.S. Fennell, 2016. An Overview of Advances in Biomass Gasification. Energy Environ. Sci., 9,
827   2939--2977
828
829   [19]   M. Said, L. Cassayre, J.-L. Dirion A. Nzihou, and X. Joulia, 2018. Influence of Nickel on Biomass
830   Pyro-Gasification: Coupled Thermodynamic and Experimental Investigations Ind. Eng. Chem.
831   Res. 57, 9788−9797
832
833   [20]   J.N. Semaan, M. Huron and E. Daouk, 2021. Pilot Scale Pyro-Gasification of Biomass and
834   Waste: Char   Characterization. *Biomass   Conv.   Bioref.* https://doi.org/10.1007/s13399-020-
835   01181-3
836
837   [21]   X. Xu, E. Jiang, M. Wang, Y. Xu, 2015. Dry and Steam Reforming of Biomass Pyrolysis Gas for
838   Rich Hydrogen Gas. Biomass and Bioenergy, 78, 6 -16
839
840   [22]   K. Akubo, M. A. Nahil, and P. T. Williams, 2019. Pyrolysis-Catalytic Steam Reforming of
841   Agricultural Biomass Wastes and Biomass Components for Production of Hydrogen/Syngas. J.
842   Energy Institute 92, 1987 - 1996
843
844   [23]   M.-J. Park, J.-H. Kim, Y.-H. Lee, H.-M. Kim, D.-W. Jeong, 2020. System Optimization for
845   Effective Hydrogen Production Via Anaerobic Digestion and Biogas Steam Reforming. Int. J.
846   hydrogen energy, 45, 30188 - 30200.
847
848   [24]   C.E. Tuna, J.L. Silveira, M.E. da Silva, R.M. Boloy, L.B. Braga, N.P. Perez, 2018. Biogas Steam
849   Reformer for Hydrogen Production: Evaluation of the Reformer Prototype and Catalysts, Int. J.
850   hydrogen energy, 43, 2108 -2120
851
852   [25]   J.A. Onwudili, 2014. Hydrothermal Gasification of Biomass for Hydrogen Production. In: Jin
853   F. (eds) Application of Hydrothermal Reactions to Biomass Conversion. Green Chemistry
854   and Sustainable Technology. Springer, Berlin, Heidelberg. https://doi.org/10.1007/978-3-
855   642-54458-3_10
856
857   [26]   D.C. Elliott, G.G. Neuenschwander, and T.R. Hart, 2018. Combined hydrothermal liquefaction
858   and catalytic hydrothermal gasification system and process for conversion of biomass
859   feedstocks. United States: N. p., 2018. Web. https://www.osti.gov/biblio/1495326-combined-
860   hydrothermal-liquefaction-catalytic-hydrothermal-gasification-system-process-conversion-
861   biomass-feedstocks
862
863   [27]   C. Yang, S. Wang, J. Yang, D. Xu, Y. Li, J. Li and Y. Zhang, 2020. Hydrothermal Liquefaction
864   and Gasification of Biomass and Model Compounds: A Review. Green Chem., 22, 8210 - 8232

[28] A. Molino, S. Chianese, D. Musmarra, 2016. Biomass Gasification Technology: the State of the Art Overview. J. Energy Chem. 25, 10−25.

[29] S. Mishra and R. Kumar Upadhyay, 2021. Review on Biomass Gasification: Gasifiers, Gasifying Mediums, and Operational Parameters. Mat. Sci. Energy Tech. 4, 329–340

[30] J. Ren, J. P. Cao, X.Y. Zhao, F.L. Yang, X.Y. Wei, 2019. Recent Advances in Syngas Production from Biomass Catalytic Gasification: A Critical Review on Reactors, Catalysts, Catalytic Mechanisms and Mathematical Models. Ren. Sust. Energy Rev. 116, 109426 - 109450.

[31] N. Couto, A. Rouboa, V. Silva, E. Monteiro, K. Bouziane, 2013. Influence of the Biomass Gasification Processes on the Final Composition of Syngas. Energy Procedia, 36, 596-606.

[32] M. Binder, M. Kraussler, M. Kuba and M. Luisser, 2019. Hydrogen from Biomass Gasification. https://www.ieabioenergy.com/wpcontent/uploads/2019/01/Wasserstoffstudie_IEA-final.pdf

[33] G. Di Marcoberardino, S. Foresti, M. Binotti and G. Manzolini. 2018. Potentiality of a Biogas Membrane Reformer for Decentralized Hydrogen Production. Chem. Eng. Process, 129, 131–141

[34] GRDF, 2022. Pyro-Gasification: a Technology with Great Potential for Producing Green Gas. https://www.grdf.fr/english/what-we-do/renewable-gases/pyro-gasification

[35] M. Elmously, n. Jäger, J. Neidel, A. Apfelbacher, and A. Hornung, 2019. Upscaling of Thermo-Catalytic Reforming Process from Lab to Pilot Scale', Ind. Eng. Chem. Res., 58 (35),15853-15862.

[36] G.V. Hecke, 2020. Private Communications with Van Hool, Birmingham, England, United Kingdom

[37] Toddington, S. 2022. Official Communication with Transport for West Midlands, Birmingham, England, United Kingdom.

[38] F.S. Lau, D.A. Bowen, R. Dihu, S. Doong, E.E. Hughes, R. Remick, R. Slimane, R. S.Q. Turn, R. Zabransky, 2002. Techno-Economic Analysis of Hydrogen Production by Gasification of Biomass Final Technical Report Work Performed Under DOE Contract Number: DE-FC36-01GO11089. Online: https://www.osti.gov/servlets/purl/816024. [Accessed: 06/03/2021]

[39] J. Yao, M. Kraussler, F. Benedikt, H. Hofbauer, 2017. Techno-economic Assessment of Hydrogen Production based on Dual Fluidized Bed Biomass Steam Gasification, Biogas Steam Reforming, and Alkaline Water Electrolysis Processes. Energy Conv. Manage., 145, 278 - 292.

[40] C. Cluzel, and A. Hope-Morley, 2016. West Midlands Low Emission Bus Delivery Plan: As study commissioned by Centro. Network West Midlands. Online: https://www.wmca.org.uk/media/1366/west-midlands-low-emission-delivery-plan_elementenergy-for-transport-for-west-midlands_july2016.pdf. [Accessed: 10/06/2020]

[41] U. Arena, and M.L. Mastellone, 2005. Pyrolysis and Gasification of Solid Wastes in Fluidised Bed Reactors. Proc. of Industrial Fluidization South Africa 53 - 68.

916   [42]   C. Wu, Q. Huang, M. Sui, Y. Yan, F. Wang, 2008. Hydrogen Production via Catalytic Steam
917   Reforming of Fast Pyrolysis Bio-oil in a Two-stage Fixed Bed Reactor System. Fuel Process
918   Technol., 89 (12), 1306-1316
919
920   [43]   A.V. Bridgwater, 1999. An Overview of Fast Pyrolysis of Biomass. Organic Geochemistry. 30,
921   1479 - 1493.
922
923   [44]   T.A. Milne, C.C. Elam, and R.J. Evans, 2002. Hydrogen from Biomass: State of the Art and
924   Research Challenges. National Renewable Energy Laboratory, Colorado, USA. Online:
925   https://www.nrel.gov/docs/legosti/old/36262.pdf. [Accessed: 20/04/2021]
926
927   [45]   Biogas Renewable Energy, 2021. Biogas Composition. Online: https://www.biogas-renewable-
928   energy.info/biogas_composition.html. [Accessed: 21/06/2021]
929
930   [46]   C.M, Kalamaras, and A.M. Efstathiou, 2013. Hydrogen Production Technologies: Current State
931   and Future Developments. Conference Papers in Science, https://doi.org/10.1155/2013/690627
932
933   [47]   Forest Research. UK, 2020. Wood Production and Trade: 2020 Provisional Figures. Online:
934   https://www.forestresearch.gov.uk/documents/8032/ukwpt21.pdf. [Accessed: 14/06/2021]
935
936   [48]   H.K. Jeswani, G. Figueroa-Torres and A. Azapagic,, 2021. The Extent of Food Waste Generation
937   in the UK and its Environmental Impacts. Finnveden, G (ed.). Sustainable Production and
938   Consumption. 26, 532 - 547.
939
940   [49]   M. Ringer, V. Putsche, and J. Scahill, 2006. Large-Scale Pyrolysis Oil Production and Economic
941   Analysis; Technical Report NREL/TP-510–37779; National Renewable Energy Laboratory, Cole
942   Boulevard, CO, USA. Online: https://www.nrel.gov/docs/fy07osti/37779.pdf. [Accessed:
943   01/05/2021]
944
945   [50]   J. Rusín, K. Kasakov and K. Chamradova, 2018. High-solids Semi-continuous Anaerobic
946   Digestion of Corn Silage in Bag-type Digester. Green Proc. Synthesis, 7(3), 268-276.
947
948   [51]   G.M. Sobamowo and S.J Ojolo, 2018. Techno-Economic Analysis of Biomass Energy Utilisation
949   through Gasification Technology for Sustainable Energy Production and Economic Development
950   in Nigeria. J. Energy (Open Access). Online: https://doi.org/10.1155/2018/4860252.
951
952   [52]   R.K. Sinnott and G. Towler, 2009. Chemical Engineering Design: Coulson and Richardson's
953   Chemical Engineering Series, Fifth Edition. Elsevier, Oxford, United Kingdom
954
955   [53]   W. Liu, J. Wang, T.L. Richard, D.S. Hartley, S. Spatari and T.A. Volk, 2017. Economic and Life
956   Cycle Assessments of Biomass Utilization for Bioenergy Products. Biofuels, Bioproducts and
957   Biorefining, 11(4) 633-647
958
959   [54]   C. Hepburn, E. Adlen, J. Beddington, E.M. Carter, S. Fuss, N.M. Dowell, J.C. Minx, P. Smith and
960   C.K. Williams, 2019. The technological and economic prospects for CO2 utilization and removal.
961   Nature, 575 (7781), 87-97
962
963   [55]   D.E. Garrett, 2012. Chemical engineering economics. Springer Science & Business Media.
964   Online: https://www.springer.com/gp/book/9789401165464. [Accessed: 20/06/2021]
965
966   [56]   Fred Economic Data, 2020. Producer price index by industry: Oil and gas field machinery and
967   equipment manufacturing, 2020. https://fred.stlouisfed.org/series/PCU333132333132.

[57] G. Towler and R. Sinnott, 2013. Chemical Engineering Design: Principles, Practice and Economics of Plant and Process Design. 2nd ed. Boston: Elsevier.

[58] H. Ritchie and M. Roser, 2007. United Kingdom: CO_2 Country Profile. Our World in Data. Online: https://ourworldindata.org/co2/country/united-kingdom?country=~GBR.] [Accessed: 11/02/2021.

[59] Q. Yi., F. Qi., B. Xiao., Z. Hu and S. Liu, 2013. Co-Firing Ramie Residue with Supplementary Coal in a Cyclone Furnace. Bioresources 8(1), 844-854

[60] D.R. Woods, 2007, Rules of Thumb in Engineering Practice. Appendix D: Capital Cost Guidelines. Online: https://onlinelibrary.wiley.com/doi/book/10.1002/9783527611119. [Accessed: 14/05/2021]

[61] Nexant Inc., 2006. Equipment Design and Cost Estimation for Small Modular Biomass Systems, Synthesis Gas Clean up and Oxygen Separation Equipment. Task 1: Cost Estimates of Small Modular Systems. National Renewable Energy Laboratory. Online: https://www.nrel.gov/docs/fy06osti/39943.pdf. [Accessed: 14/03/2021]

[62] J.M. Arati, 2009. Evaluating the economic feasibility of Anaerobic Digestion of Kawangware Market Waste. Master's Dissertation, Kansas State University, USA. Online: https://krex.k-state.edu/dspace/handle/2097/2200. [Accessed: 02/03/2021]

[63] Y.K. Salkuyeh, B.A. Saville and H.L. MacLean, 2018. Techno-economic Analysis and Life Cycle Assessment of Hydrogen Production from Different Biomass Gasification Processes. Int. J. Hydrogen Energy 43(20), 9514-9528

[64] A. Mustafa, R.K. Calay and M.Y. Mustafa, 2017. A Techno-Economic Study of a Biomass Gasification Plant for the Production of Transport Biofuel for Small Communities. Energy Procedia 112, 529 – 536

[65] Y. Ji-chao and B. Sobhani, 2021. Integration of biomass gasification with a supercritical CO_2 and Kalina cycles in a combined heating and power system: A thermodynamic and exergoeconomic analysis. Energy 222, 119980-119995.

[66] L. Collins, 2020. Green Hydrogen on Sale in Open Market at 80% higher price than Grey H_2. Recharge News. Online: https://www.rechargenews.com/transition/green-hydrogen-on-sale-in-open-market-at-80-higher-price-than-grey-h2/2-1-743348. [Accessed: 11/02/2021]

[67] F. Calise, M.D. D'Accadia, M. Santarelli, A. Lanzini and D. Ferrero, 2019. Solar Hydrogen Production: Processes, Systems and Technologies. Academic Press.

[68] K.S. Shiva K.S. and V. Himabindu, 2019. Hydrogen Production by PEM Water Electrolysis – A Review. Mater. Sci. Energy Technol. 2, 442–454

[69] Z. Xin-gang, J. Gui-wu, L. Ang, and L. Yun, 2016. Technology, Cost and Performance of Waste-to-Energy Incineration Industry in China. Renewable and Sustainable Energy Reviews 55, 115–130.

[70] J. Eke and J.A. Onwudili, 2021. Economic evaluation of a Hypothetical Integrated Energy Recovery System for Trommel Fines. Waste Management 124, 213-223

1020   [71]   G. Chen, J. Andries, Z. Luo and H. Spliethoff, 2003. Biomass Pyrolysis/Gasification for Product
1021   Gas Production: the Overall Investigation of Parametric Effect. Energy Conv. Manage. 44, 1875–
1022   1884
1023
1024
1025
1026
1027

Declaration of interests

☒ 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.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: