

ADVANCED THERMOCHEMICAL CONVERSION OF VARIOUS WASTE FEEDSTOCK WITH CCS FOR HYDROGEN PRODUCTION – A LIFE CYCLE ASSESSMENT

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ABSTRACT: A life cycle assessment on a process that converts Municipal Solid Waste (MSW) to Hydrogen via Gasification at scale is presented. The produced hydrogen meets specification for fuel cells in transport applications. Crucially, the process employs the capture and permanent geological sequestration of direct carbon dioxide emissions from the transformation of waste. The process is considered a negative emissions technology i.e. Bioenergy with Carbon Capture and Storage (BECCS), with these capabilities highly dependent on the biogenic carbon fraction of the waste feedstock. The gasification process is compared to other low-carbon hydrogen alternative such as steam methane reforming and autothermal reforming with CCS and electrolysis of water.

Keywords: waste, hydrogen, gasification, CO₂ reduction, life cycle assessment

1 INTRODUCTION

Hydrogen produced from low-carbon routes will play a crucial and complementary role to green electrification in reaching net zero. It will particularly be useful in sectors where electrification is not feasible, for example, in industrial furnaces, long distance and heavy-duty transport. It may also be used for energy storage and flexible power generation. Low-carbon hydrogen is expected to make up 20-35 percent of UK's final energy consumption by 2050, which is a hydrogen deployment of 250-460 TWh [1].

Currently, 94% of hydrogen worldwide is produced from steam methane reformation of natural gas or gasified coal with a primary application as an industrial feedstock in refineries and ammonia production. This route, so-called Grey-H₂, produces approximately 11 and 13 kg CO₂ eq./kg H₂ [2]. Thus, there is a need to transition to production routes that are compatible with a low-carbon economy. One such route is, Blue-H₂, which is effectively natural gas gasification (Grey-H₂) coupled with carbon capture and storage. This route can be implemented via either types Steam methane reforming (SMR) or auto thermal reforming (ATR). While current SMR plants may be retrofitted with CCS in a relatively straightforward way, ATR is capable of reaching greater carbon capture rates owing to a process that produces a single stream of concentrated CO₂. Overall, Blue-H₂ benefits from existing infrastructure, commercial, mature technologies, thus providing an attractive medium-term alternative, restricted by economics and technical efficiency of CCS technologies.

The small remaining percentage of current worldwide hydrogen is produced by electrolysis with commercial scale seeing limited adoption until recently due to high capital costs (~3 and 15 €/kg H₂ produced, electrolyser-size dependent) [3]. Due to high electricity demands to electrochemically split water, this technology is considered a low-carbon technology (Green-H₂) only when electricity is supplied from renewable sources, such as solar and wind. Early adopters of Green-H₂ are small-scale systems for hydrogen mobility projects, while larger, centralised projects will see a steady ramp-up from 2030 onwards.

Biohydrogen (Bio-H₂) with carbon capture and storage, herein termed Bio-H₂, is the production of H₂ by thermal conversion of biomass or other organic materials via gasification or pyrolysis. Although innovation and further development is required for commercial

deployment of this technology compared to Blue- and Green-H₂, Bio-H₂ coupled with CCS (BECCS) can make a significant contribution to meeting 2050 climate change targets as they support decarbonisation of problematic sectors through its carbon-negative technology capabilities [4], [5]. Extensive research has been conducted on biomass gasification. The process can be more attractive when the feedstock considered is waste, resulting in lower costs and added environmental benefits, diverting waste from current polluting alternatives (e.g. landfill and incineration) [6].

This work aims to understand the strengths and challenges of on-site electrolysis (Green-H₂), SMR and ATR with CCS (Blue-H₂) and gasification of biomass-containing feedstock (Bio-H₂) from an environmental perspective to better support decision making and deployment in the future.

2 TECHNOLOGICAL ASPECTS

2.1 Process description: Waste-to-H₂

The Waste-to-H₂ plant design is based on existing advanced demonstration plant close to commercialization. The feedstocks analysed were Municipal Solid Waste (MSW) and Waste wood (WW) (Table I). The process employs a steam-oxygen blown fluidized bed gasifier (700-800°C) and a thermal plasma-powered tar-reformer (1200°C). Dry filters, acid scrubbers, and alkali scrubbers are used to remove contaminants like heavy metals, sulfur, and chlorine. Water gas shift reactors increase the concentration of H₂ and CO₂. The purified gas is fed into a pre-combustion carbon capture unit using Monoethanolamine (MEA) solvent. CO₂ is absorbed in the absorber and stripped using steam in the stripper. The system achieves 90% carbon capture removal rate. The remaining product gas is processed to obtain high-purity hydrogen which is pressurized at 200 bar. Remaining tail gas is used to generate electricity via a Jenbacher gas engine. The dehydrated and compressed CO₂, maintained at a pressure of 60 bar, undergoes transportation to the nearest carbon capture and utilization (CCUS) cluster and involves lorry, shipping tankers and pipelines, eventually reaching the North Sea and injected into a deep saline aquifer for long-term storage. To compensate for pressure drops during pipeline transportation, the CO₂ is re-pressurized from 60 bar to 120 bar.

Table I: Composition of waste feedstock

Proximate analysis [wt%, as received]	RDF	Waste Wood
Fixed Carbon	8.90	10.75
Volatile matter	64.70	64.24
Ash	11.80	0.41
Moisture	14.60	24.6
Ultimate analysis [wt%, dry ash free (DAF)]	RDF	Waste Wood
Fossil Carbon	20.51	0.80
Biogenic Carbon	36.23	50.13
Hydrogen	6.86	5.76
Oxygen	31.78	43.01
Nitrogen	4.1	0.28
Sulphur	0.18	0.01
Chlorine	0.34	0.01

2.2 CO₂ capture technologies

Pre- and post-combustion capture technologies are two different approaches used to reduce carbon emissions from power generation and industrial processes. Post-combustion capture aims to capture and separate CO₂ from flue gas streams after the combustion process. It has been successfully applied in the power sector, with the Petra Nova Carbon Capture project and the Boundary Dam Carbon Capture project, which are both coal-fired plants retrofitted with CCUS [7]. It is also being deployed in the waste-to-energy sector, for example, the Klemetsrud Waste-to-Energy Plant in Oslo, Norway plans to implement a full-scale amine-scrubbing based post-combustion capture [8].

In the Bio-H₂ gasification process and Blue-H₂ steam methane reforming process presented here, a pre-combustion capture technology is considered as it allows CO₂ to be captured from the syngas after the shift reactors. This is typically more efficient as the stream is CO₂-rich and at higher pressures than if captured from a flue-gas stream.

2.3 Hydrogen

Hydrogen in the UK is expected to find use in industrial heating and in fuel cells, with different purity requirements. For residential/commercial combustion appliances (e.g. boilers, cookers and similar applications), a purity of $\geq 98.0\%$ is required while industrial fuel for power generation and heat generation require $\geq 99.9\%$ purity [9]. Fuel quality specifications for fuel cell road vehicle and stationary applications are more stringent [9]. For road vehicles, the fuel cell that is expected to dominate the market are proton exchange membrane (PEM) fuel cells with a hydrogen purity of $\geq 99.97\%$. The alternative fuel cells, namely Molten Carbonate Fuel Cells (MCFC) and Solid oxide fuel cells (SOFC), are expected to withstand lower purity levels for hydrogen as the high temperatures ($>650\text{ }^\circ\text{C}$) and catalytic nature of these FCs prevent CO poisoning [10]. The current purification technology capable of reaching such high purities for PEM-FC is pressure swing adsorption (PSA).

Distribution options for hydrogen to their respective hydrogen refuelling stations include of liquid hydrogen by truck, compressed H₂ by truck or by gas network pipelines. To liquefy for truck transport, hydrogen is compressed to 80 bar followed by cryogenic cooling and will require specialised liquefied hydrogen tankers for transport. BOC compressed hydrogen trucks are compressed to 280 bar.

The transportation of hydrogen via pipeline in the UK is demonstrated with a blend of up to 20% (vol.) hydrogen in the gas [11]. The Fife H100 project is looking at supplying 100% through a purpose-built gas network from 2022 [12]. Thus, transport of hydrogen by pipeline will take place in the medium to long term with natural gas blended phases with a full conversion of the gas network expected in 2040s [13]. Injection in the grid will be at low pressures, while dispensing of H₂ into fuel cell cars will require higher pressures. Hydrogen is expected collect impurities and thus lose purity during gas network delivery and will require repurification at a hydrogen refuelling station (HRS). For bulk storage of hydrogen, salt caverns have been identified, but is out of scope for this study [14].

Recent research has shown that hydrogen leaks to the atmosphere during transport, storage and dispensing are expected to impart an indirect greenhouse gas effect. Considered an indirect greenhouse gas, oxidation of H₂ competes with the primary oxidation pathway of methane which results in longer methane lifetime, thereby contributing to global warming [15]. This aspect, previously, not explored at length will be analysed.

3 METHODOLOGY

3.1 LCA Methodology

The study adheres to ISO 14040 and ISO 14044 guidelines and is based on GaBi 10.0.0.71 using Thinkstep and ecoinvent v3.6 databases [16], [17]. Primary data for Bio-H₂ is obtained through ASPEN Plus modeling and supplemented with plant data from a waste gasification company. An attributional life cycle assessment (LCA) is conducted for biohydrogen from waste wood and MSW and corresponding climate change hotspots analysed. Zero-burden approach for waste is considered. Life cycle impacts were assessed across the categories that represent the highest environmental priorities according to normalisation using the EF 3.0 global reference normalisation and weighting factors [18]. Hauschild et al. provides a detailed description of these impact categories [19]. Electricity from tail gas replaces UK grid mix and recovery of metals from MSW for RDF preparation are considered. The system boundary for the hotspot analysis includes plant construction, CO₂ transport and storage and fugitive hydrogen emissions.

Bio-H₂ is then compared to alternative hydrogen production routes. Data for Blue-H₂ via SMR and ATR is based on 90% amine-based capture modelled by Antonini et al. (2020). Green-H₂ is based on a liquid alkaline based electrolyser stack with an efficiency of at 62.5%, approximated the CUTE project [21] and is supplied by either solar or offshore wind. System boundary is depicted in Figure II. Further inventory data information can be found in Amaya-Santos et al. (2021) and Chari et al. (2023).

The functional unit is the production of 1 MW_{HHV} of transport-grade H₂ ($>99.97\%$ purity) compressed to 200 bar for fuel cell dispensing.

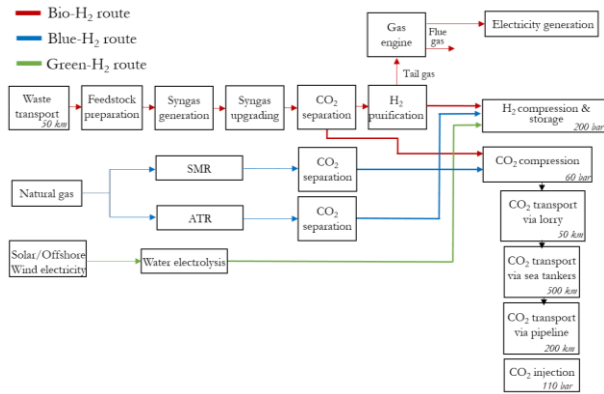


Figure 2: System boundary

3.2 Fugitive H₂ emissions

With recent atmospheric science literature pointing towards the indirect GHG impact of hydrogen, a preliminary analysis on fugitive H₂ emissions only during transport via pipeline was considered. For hydrogen distribution, transport via pipeline followed by dispensing at 200 bar was considered; re-purification of hydrogen at HRS via PSA and a blended natural gas scenarios were not included as the focus was determining the magnitude and impact of fugitive H₂ emissions.

BEIS reports detailed emissions across the hydrogen supply chain, amounting on average a to 1-10% leakage rate. Table II details the leakage rates specific to pipeline transport relevant to this work. Uncertainties for leakage estimates are still high and are expected to reduce as infrastructure develops. For example, the replacing the majority of the iron-piped gas network with polyethylene pipes would reduce fugitive emissions [24].

Table II: Hydrogen leakage rates during pipeline transportation (adapted from Frazer-Nash Consultancy (2022))

Transport leakage rate (%)	Predicted emission confidence level	
	50%	99%
National transmission system	0.04	0.48
Distribution network	0.26	0.53

The atmospheric models for estimating global warming potential for hydrogen based on natural hydrogen sources and sinks, still shows large deviation in literature. Although not yet asserted in IPCC, an average GWP over 100-year timescale for hydrogen was calculated using values in Table III.

Table III: 100-yr GWP potential of hydrogen

Source	100-yr GWP (kg CO ₂ eq.)	Uncertainty (+/- kg CO ₂ eq.)
Warwick et al. (2022)	11	5
Derwent et al., (2020)	5	1
Field & Derwent, (2021)	3.3	1.4

4 RESULTS

4.1 Climate change hotspot

The total climate change impact for MSW and WW feedstock is negative, owing to the capture and permanent sequestration of biogenic CO₂ as shown in Figure III. The 100% biogenic content of WW compared to 64% for MSW, makes the WW process more carbon negative. However, WW does exhibit a greater positive impact, on the process side, due to lower calorific value and thus greater feed throughput. WW also generates more equivalent CO₂ which when sequestered creates further savings. The credits associated to biogenic C sequestration has a total climate change impact of -108 and -288 kg CO₂ eq./ FU for WW and MSW respectively thus showing a high sensitivity to changes in biogenic/fossil carbon content of the waste. If biogenic carbon content and/or carbon capture rate were to increase, the process will return greater carbon negative capabilities. Credits associated to recovery of ferrous and non-ferrous metals in MSW also provide savings of around 6%.

The stripper solvent regeneration unit (carbon capture section) constitutes the most thermal energy intensive unit even with some thermal energy recovered in other stages used to offset this demand. The contribution from carbon capture is about 8.8% and 10.1% for WW and MSW. Current amine solvent-based carbon capture technology presented here study could be substituted with other established technologies like Selexol, Rectisol, or Benfield. This substitution has the potential to enhance energy savings and should be explored.

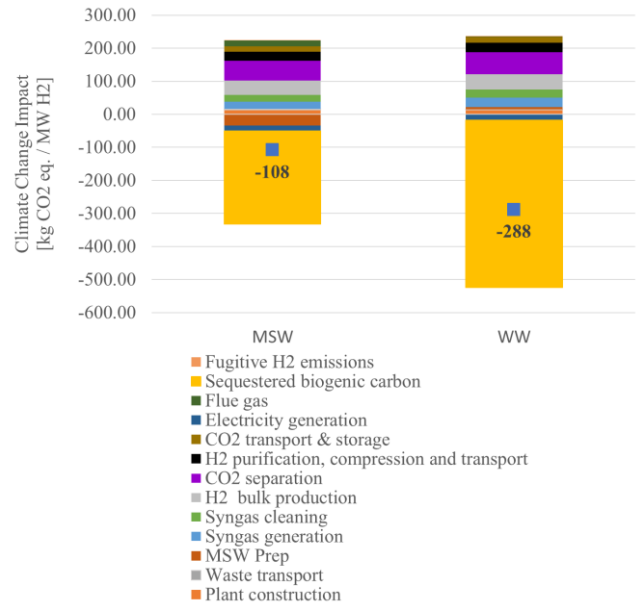


Figure 3: Climate change impact hotspot for Biohydrogen from WW and MSW

Waste transport carries negligible (<1%) impact contribution primarily due to a short transport distance and lower mass carrying load (compared to e.g. CO₂ transport). While CO₂ transport and storage is 2-3% which is dominated by transport via lorry despite it being used over the shortest distance. Thus, it is worth highlighting the equivalent impacts between the different modes of transport used. For example, climate change impacts for transport via lorry, sea tankers and pipeline are 0.16, 0.01

and 0.0001 kg CO₂ eq. / tkm. The UK is well poised to exploit infrastructure and expertise of its expansive gas network and transport waterways [29]. Transportation mode and distances will be a useful optimisation key specific to each industrial case.

There is large uncertainty around the magnitude and impact of the fugitive emissions. However, the impact is insignificant with 1.1 kg CO₂ eq./MW H₂. Considering a large scale shift towards hydrogen systems, fugitive emissions should continue to be investigated.

4.2 Climate change impact of alternative low-carbon hydrogen technologies

The alternative low-carbon hydrogen technologies explored include Bio-H₂ via waste gasification, Blue-H₂ via SMR/ATR and Green-H₂ via electrolysis. Most greenhouse gas emissions from these technologies are dominated by the core process or feedstock/electricity source, shown in Figure IV.

As of 2020, no SMR or ATR specific plants with CCS capabilities were operational, however, For Blue-H₂ via SMR/ATR, 30-40% of emissions arise from the natural gas feedstock itself. Changes to the source of natural gas would thus influence results for example, LNG vs gas pipeline proportion of imports. In addition, carbon footprint of LNG imports to the UK could improve, for example through decarbonisation of shipping. The conversion process dominates climate change impact with ~60% with ATR showing a lower impact due to greater CCS energy efficiency (MJ/kg CO₂ captured). This improved CO₂ capture efficiency for ATR, brings the process similar in impact to the Green-H₂ via photovoltaic energy.

Currently only small on-site electrolyzers are used for hydrogen production. In 2030 and 2035, large scale centralized electrolyzers will begin deployment in the UK. [1] Electrolyzers require electricity a magnitude greater than the other chemical conversion processes to hydrogen analysed. Thus, current electrolyzers fed by the current grid mix supply render the process highly carbon intensive. This is expected to change as grid carbon intensity reduces with shifts to more renewable energy sources. For Green-H₂, system transformation towards renewables is crucial as it cannot be considered low carbon electricity if it diverts supply of renewables from other demands. Climate change impact when electrolysis is supplied by exclusively solar or offshore wind are 99 and 23 kg CO₂ eq/ MW H₂ in this study. Green-H₂ via solar is equivalent to Blue-H₂ via ATR due to the several energy intensive stages during manufacturing of mono- and multi-crystalline silicon solar cells. Similarly, for Green-H₂ via off-shore wind, manufacturing and construction stages of wind turbines dominate most environmental impacts, primarily from the manufacture of steel and cement. These results may change as the manufacturing processes turn green much in the same way as the grid-electricity – with energy for solar cell manufacture sourced from renewables and with sustainable improvements in steel and cement industries. Additionally, improved end-of-life and recycling of solar cells and wind turbines would improve environmental impacts of these technologies. The study focused on alkaline electrolyzers, however, many electrolyser technologies (e.g. PEM) exist and should be explored further.

The Bio-H₂ pathway is the only carbon-negative hydrogen technology due to sequestration of biogenic carbon in waste feedstock. The positive climate change

impacts for Bio-H₂ are higher or equivalent to Blue-H₂ via SMR, arising mainly from the feedstock conversion to high-purity hydrogen and carbon capture. These findings highlight the potential of Bio-H₂ as a promising method for waste disposal while simultaneously serving as a viable carbon-negative technology [30].

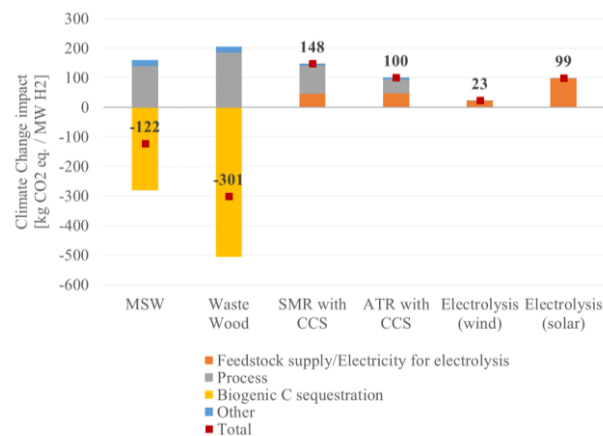


Figure 4: Climate change impact of alternative low carbon hydrogen production routes

4.3 Additional environmental impacts of alternative low-carbon hydrogen technologies

Although international governmental attention is heavily placed on abating climate change by reducing greenhouse gas emissions to the environment, this must be balanced by an equal investigation into other environmental impacts. The importance of other environmental categories is highlighted in the planetary boundary framework by Rockström et al. (2009) which states "transgressing one or more planetary boundaries may be deleterious or even catastrophic due to the risk of crossing thresholds that will trigger non-linear, abrupt environmental change within continental-scale to planetary-scale systems.". Aside from climate change, a few of these boundaries, for example, biodiversity integrity and biogeochemical flows, have already been transgressed to points of high uncertainty and thus incalculable risk [32].

In this context, these hydrogen production routes have also been compared across other environmental categories in Figure V. What is evident is that no one technology is ranked most environmentally advantageous across categories. Bio-H₂ although extremely crucial for climate change abatement with its carbon negative capabilities, underperforms in other categories owing to the relatively nascent (compared to SMR/ATR) processing technologies. Using waste as a feedstock also involves further cleaning stages due to its heterogeneous nature, resulting in larger impacts in categories such as Ecotoxicity and Eutrophication. Blue-H₂ has a greater impact on those categories that are related to depletion of fossil resources. Green-H₂ with solar more negatively impacts Acidification due to higher proportion of SO₂ emissions in silicon solar cell manufacture and similarly, Green-H₂ with wind due to materials used in construction of wind turbines that emit a higher proportion of NO₂ [33], [34]. Also, electrolysis is expected to require large amounts of freshwater (or seawater coupled with desalination process) that can impact water scarcity [35].

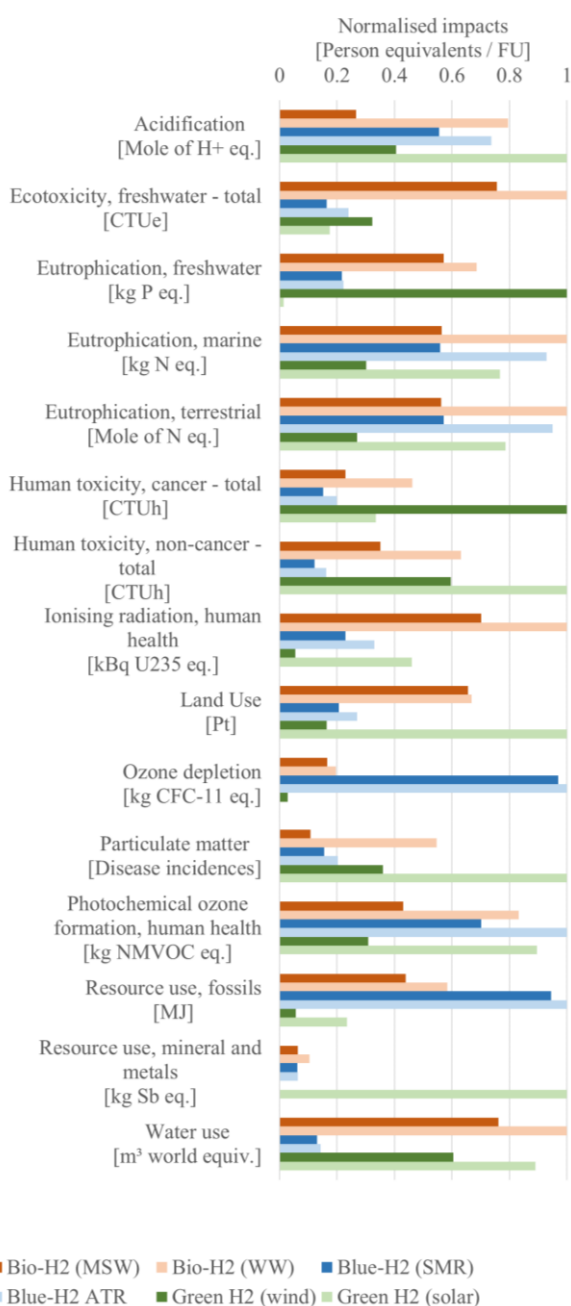


Figure 5: Normalised impacts of all other impact categories for low-carbon H₂ production routes

5 CONCLUSION

Bio-H₂ is proposed as a complementary technology to aid the near- and medium-term transition as well as a long-term complement to other low-carbon production routes. Further to the inherent advantages of clean hydrogen as a product, a process that converts solid waste to hydrogen has the major advantages of diverting waste from landfill, and producing a relatively pure stream of CO₂ ready for storage. However other obstacles are present, such as the limited feedstock availability and the technological risks associated with waste gasification and pre-combustion CCS.

Future healthy and dynamic hydrogen market will include multiple sources and end-uses for hydrogen.

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