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A critical review on the principles, applications, and challenges of waste-to-hydrogen technologies

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ABSTRACT

Hydrogen sourced from energy recovery processes and conversion of waste materials is a method of providing both a clean fuel and a sustainable waste management alternative to landfill and incineration. The question is whether waste-to-hydrogen can become part of the zero-carbon future energy mix and serve as one of the cleaner hydrogen sources which is economically viable and environmentally friendly. This work critically assessed the potential of waste as a source of hydrogen production via various thermochemical (gasification and pyrolysis) and biochemical (fermentation and photolysis) processes. Research has shown hydrogen production yields of 33.6 mol/kg and hydrogen concentrations of 82% from mixed waste feedstock gasification. Biochemical methods such as fermentation can produce hydrogen up to 418.6 mL/g. Factors including feedstock quality, process requirements and technology availability were reviewed to guide technology selection and system design. Current technology status and bottlenecks were discussed to shape future development priorities. These bottlenecks include expensive production and operation processes, heterogeneous feedstock, low process efficiencies, inadequate management and logistics, and lack of policy support. Improvements to hydrogen yields and production rates are related to feedstock processing and advanced energy efficiency processes such as torrefaction of feedstock which has shown thermal efficiency of gasification up to 4 MJ/kg. This will affect the economic feasibility and concerns around required improvements to bring the costs down to allow waste to be viewed as a serious competitor for hydrogen production. Recommendations were also made for financially competitive waste-to-hydrogen development to be part of a combined solution for future energy needs.

1. Introduction

Waste-to-Hydrogen (WtH) is a specific division of the Energy from Waste (EfW) concept with an emphasis on hydrogen production from waste materials. The potential of hydrogen as a fuel is based on its properties and the interest from industry and governments for a clean fuel to replace fossil fuels. Energy consumption is the main contributor to CO₂ emissions, and global CO₂ emissions from the combustion of fossil fuels reached 33.1 Gt in 2018 [1]. Finding an alternative to fossil fuels will, therefore, be an important step in reducing global CO₂ emissions. Hydrogen is a clean fuel which produces water when combusted or used in an electrolyser and has the potential to contribute to the decarbonisation of the energy sector if hydrogen production is based on renewable energy sources. However, around 96% hydrogen is

currently produced by the transformation of fossil fuels (48% from natural gas, 30% from heavy oils and naphtha, and 18% from coal), which leads to an annual CO₂ production of 560 million tonnes, equivalent to 1.7% of the global energy-related CO₂ emissions [1]. To achieve the full benefits of hydrogen as a clean, versatile, and efficient fuel, it is essential renewable energy sources are utilised.

Conventional hydrogen production technologies include steam reforming of natural gas (methane) and petroleum, catalytic decomposition of natural gas, partial oxidation of heavy hydrocarbons, and gasification of coal or coke. These technologies are energy-intensive (i.e., high-temperature requirements) with 96% sourced from fossil fuels [3]. Natural gas reforming has an efficiency of 70–75% while coal gasification has a lower efficiency of 45–65%, and all fossil fuel processes create high CO₂ emissions [4]. Alternatively, thermochemical conversion of biomass, water via electrolysis, and biochemical methods

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List of abbreviations

WtH	Waste to Hydrogen
EfW	Energy from Waste
SMR	Steam Methane Reforming
H ₂	Hydrogen
CO	Carbon Monoxide
CO ₂	Carbon Dioxide
CH ₄	Methane
CaO	Calcium Oxide
S/B	Steam to Biomass ratio
ER	Equivalence Ratio
atm	atmospheres
PEMFC	Proton Exchange Membrane Fuel Cell
HSC	Hydrogen Supply Chain

LCA	Life Cycle Analysis
GWP	Global Warming Potential
GHG	Greenhouse Gases
CCS	Carbon Capture and Storage
HHP	Higher Heating Value
LHV	Lower Heating Value
MSW	Municipal Solid Waste
RDF	Refuse Derived Fuel
SRF	Solid Recovered Fuel
FW	Food Wood
VSS	Volatile Suspended Solids
VS	Volatile Solids
SOFC	Solid Oxide Fuel Cell
PSA	Pressure Swing Adsorption
CBA	Cost Benefit Analysis

involving bacteria and algae through anaerobic digestion and fermentation can be used for low-carbon hydrogen production.

Sustainable waste management is a global challenge and urgently in demand under the pressure of increasing waste production. The global waste generation will reach 6 million tonnes/day by 2025 [5], coinciding with a growing global population with a rate of 1.18% each year [6]. Sustainable waste management practices are necessary to tackle the increasing waste streams for the health, sanitation and wellbeing of the global population. Conventional waste management practices rely on landfills and incineration which have limited energy efficiencies and adverse environmental impacts. In 2012, the amount of energy lost in the EU through the use of incinerators for waste disposal was 1,805 PJ and through landfill was 1409 PJ [7]. Landfill-based waste management emits a massive amount of methane whose global warming potential is 21 times that of CO₂ over a 100-year period [8]. Solid waste landfills account for about 14% of global methane emissions [9]. Sustainable waste management can be achieved via the conversion of waste products into useful resources and commodities [11]. This will reduce the large volumes of waste entering landfill sites and reduce the associated water, soil and air pollution (methane, dioxins, leachate) [12]. Finding sustainable and efficient methods to deal with waste is on the agenda of many countries, organisations and governments including the World Energy Council [5], UN [13], European Commission [14], EEA [15], EU [16], DEFRA of the UK Government [17]. These reports look at the environmental impacts of current waste practices, economics and the potential of increased energy security through local energy production [17].

WtH contributes to developing low-carbon waste management practices which will ultimately facilitate the fulfilment of the UN's Sustainable Development Goals, e.g., Affordable and Clean Energy, and Sustainable Cities and Communities. WtH is an important global concept which has a role in the future of sustainable energy resources and reducing the impact of humans on the environment. The discussion in this article centres on the potential to utilise waste through conversion to hydrogen for further energy utilisation. This can be achieved through improving waste management schemes, hydrogen production, operations and technology, which echoes with the typical concerns in the hydrogen industry over transportation, storage, and utilisation [18].

This review paper provides an update on the current processes, bottlenecks and applications of waste-to-hydrogen technology. Also bridges the gap between studies on thermochemical and biochemical methods, with hydrogen production. Existing papers, Table 1, detail biomass, waste or MSW gasification in terms of the technology, yield and operating conditions, though few studies continue towards the issues of upscaling or envisioning the bottlenecks associated with industrial applications.

2. Hydrogen and waste

The following sections discuss the character and behaviour of hydrogen gas as the product and waste as feedstock of the waste-to-hydrogen concept.

2.1. Hydrogen

Hydrogen is present on the earth surface at 0.14% and has trace amounts in the atmosphere [19]. Hydrogen is found in compounds and formed by decomposition or conversion of other molecules such as hydrocarbons [20]. Global hydrogen production currently is about 60 million tonnes/year for ammonia production, for oil refineries to generate lighter oil fraction, chemical and pharmaceutical industries, space exploration and power generation [4].

At ambient temperature and atmospheric pressure, 1 kg of hydrogen gas has a volume of 11 m³ with a density of 0.09 kg/m³ [18]. Hydrogen has a gravimetric energy density of 141.8 MJ/kg and higher energy content per mass compared to petroleum [21]. With respect to net calorific value, 1 kg of hydrogen is equivalent to 2.75 kg of gasoline and 6 kg of methanol [2]. Hydrogen has a low boiling point of −252.9 °C, and is stored in insulated pressure vessels (e.g., high-pressure compression or low-temperature cryogenic liquefaction) [22]. Hydrogen can be combined with other materials such as metal hydride or carbon materials for solid fuel storage options [23]. Hydrogen is an efficient fuel that can make it competitive with other alternative energy sources such as wind, solar, tidal and geothermal in terms of green credentials and low CO₂ emissions.

2.2. Waste

Waste suitable for WtH includes biomass wood waste (e.g., timber or paper industry waste), municipal solid waste (MSW), sewage sludge, packaging and plastics, solid recovered fuel (SRF) and refuse derived fuel (RDF) derived from MSW. MSW normally includes biodegradable waste, recyclable material, inert waste, electronic waste, hazardous and toxic waste which is discarded by the public [24]. The average lower heating value of MSW is around 10 GJ/Mg in the EU which is low compared to fossil fuels, though can be higher (~25 GJ/Mg) with pre-treatment of the feedstock [25]. A higher LHV for feedstock combined with optimum operating conditions for the chosen conversion technology, can lead to higher hydrogen concentration in syngas. High gas yields do not always equate to high LHV, this depends on the composition of the syngas, for example high nitrogen content from using air gasifying agent will lower the HHV [129].

Most thermochemical WtH processes work on carbon-based waste and are therefore considered appropriate for food waste recycling/

Table 1
Summary of related research and papers than can be used as a comparison to this study.

Reference	Focus of study	Similar factors/parameters	Main differences between studies
Ramos et al. [12]	Biomass for fuel via co-gasification with waste, equipment, operation conditions	Review of biomass and waste as fuel	Hydrogen as a primary product not discussed
Nikolaidis and Poullikkas [18]	Review of major hydrogen production, factors and conditions from different sources	Includes biomass feedstock and thermochemical and biological processes	No focus on waste as feedstock
Baykara [19]	Overview of hydrogen; sources and production methods.	Discussion of biomass for future fuel.	No specifics on hydrogen production from waste
Dutta [21]	Review on hydrogen; clean and green production, safety and applications	Prospects for hydrogen as a fuel for climate and environmental reasons. Fuel cells and storage issues.	Mentions water waste but not issues associated
Sikarwar et al. [22]	Range of biofuels for biomass gasification to gas and liquid fuels	Hydrogen production, gasification conditions and factors. Importance of processing and syngas cleaning	Synthesis of all biofuels. Issues around hydrogen from waste not fully discussed
Lombardi et al. [25]	Review of thermochemical treatments for different wastes, plant sizes and energy recovery	Discussion on thermochemical treatments as waste management	Other forms of waste to energy. Biological methods to hydrogen not included.
Arena [37]	MSW gasification; process, operational conditions, plant configuration	Discussion on gasification for processing waste, operating parameters and environmental impact	Biological conversion technologies and hydrogen as the product not included.
Watson et al. [43]	Review of biowaste and gasification technology (reactors). Social, environmental and economic assessment.	Gasification parameters (conditions, gasification agents) and bottlenecks for waste.	Biological conversion technologies not discussed.
Parthasarathy and Narayanan [48]	Review of steam gasification of biomass to hydrogen	Factors effecting hydrogen production from biomass, steam gasification conditions	Biological conversion technologies not discussed.
Dincer and Acar [64]	Review of hydrogen production. Sustainability issues, renewable and non-renewable sources	Hydrogen production methods- thermochemical and biological. Discussion on environmental and economic factors	All hydrogen production methods discussed not focussed on waste
Materazzi et al. [106]	Biohydrogen from waste gasification. Plant design and specifications	Waste gasification, hydrogen applications, feedstock and syngas characteristics, processing	Biological conversion technologies not discussed.
Mohanty et al. [118]	Review of hydrogen from biomass. Challenges and opportunities	Gasification and pyrolysis of biomass. Biological methods;	No specifics on hydrogen production from waste

Table 1 (continued)

Reference	Focus of study	Similar factors/parameters	Main differences between studies
Valente et al. [125]	Eco-efficiency of hydrogen biomass and SMR gasification	fermentation and photolysis Environmental aspects, economic feasibility of hydrogen production	No specifics on hydrogen production from waste

valorisation though the potentially high moisture content affects the practical use and lead to energy losses [28]. Eker and Sarp [29] stated that agricultural waste is featured by high carbohydrate, nutrient and mineral contents. Ramos [12] found that the heterogeneity of MSW for gasification was a disadvantage due to low volumetric energy density which reduced the process efficiency and profitability. Dong et al. [30] suggested that the inconsistent size was a main technical problem of heterogeneity of MSW.

Biomass has a moisture content range of 30–60% (hygroscopic), low bulk density (compared to oil, coal and natural gas) and high volatile matter content (70-88 wt%) [32]. Untreated biomass has an overall low energy value and corresponds to low conversion efficiencies [31]. For the thermochemical processes, it is necessary to reduce the moisture content of feedstock as part of a pre-treatment process [33] as lower moisture content aids gasification production, improves syngas quality and energy efficiency and reduces emissions [12]. Extra energy is required for evaporation in the range of 2242 kJ/kg moisture [26]. However, studies by Hu et al. [34] and Dominguez et al. [35] suggest that high moisture content in situ gasification reactions could promote hydrogen formation reactions. For syngas production from pyrolysis, the moisture content needs to be below 30 wt%, ideally between 15 wt% and 10 wt% to reduce energy loss [31].

3. Waste to hydrogen

WtH conversion methods can be split into thermochemical and biochemical techniques differing by energy requirements, operating conditions (temperatures and pressures), feedstock inputs, efficiencies, reaction times and final yields. In general, the thermochemical processes, i.e., gasification and pyrolysis are faster than the biochemical ones (e.g., fermentation), have higher stoichiometric hydrogen yields, higher conversion efficiencies and shorter reaction times [36]. However biochemical processes are less energy intensive as they operate under moderate energy conditions leading to lower hydrogen yields (mol H₂/mol feedstock).

3.1. Thermochemical technologies

Thermochemical WtH techniques including gasification and pyrolysis are essential for a sustainable integrated MSW management system [37]. Fig. 1 is a simplified pyrolysis and gasification process diagram for hydrogen production, for illustrative and comparison purposes (stages and parameters) inspired by studies such as Iribarren et al. [33] and Arena [37]. The advantages of the thermochemical processes regarding waste management include a high reduction in waste in mass (70–80%) and volume (~80–90%) preserving landfill space [25]. Organic waste treatment via thermochemical processing before landfill leads to a reduction of landfill gas (LFG), a greenhouse gas emission comprising of CH₄ (50–60%), CO₂ (35%), and volatile organic compounds (<1%) [8]. High temperatures destroy organic contaminants such as halogenated hydrocarbons, by increasing the concentration and immobilization of inorganic contaminants thus leading to safe or useful disposal [38]. Using the solid residues of thermochemical processes such as bottom ash and slag further reduces landfill waste and increases the cost-effectiveness of the processes [37].

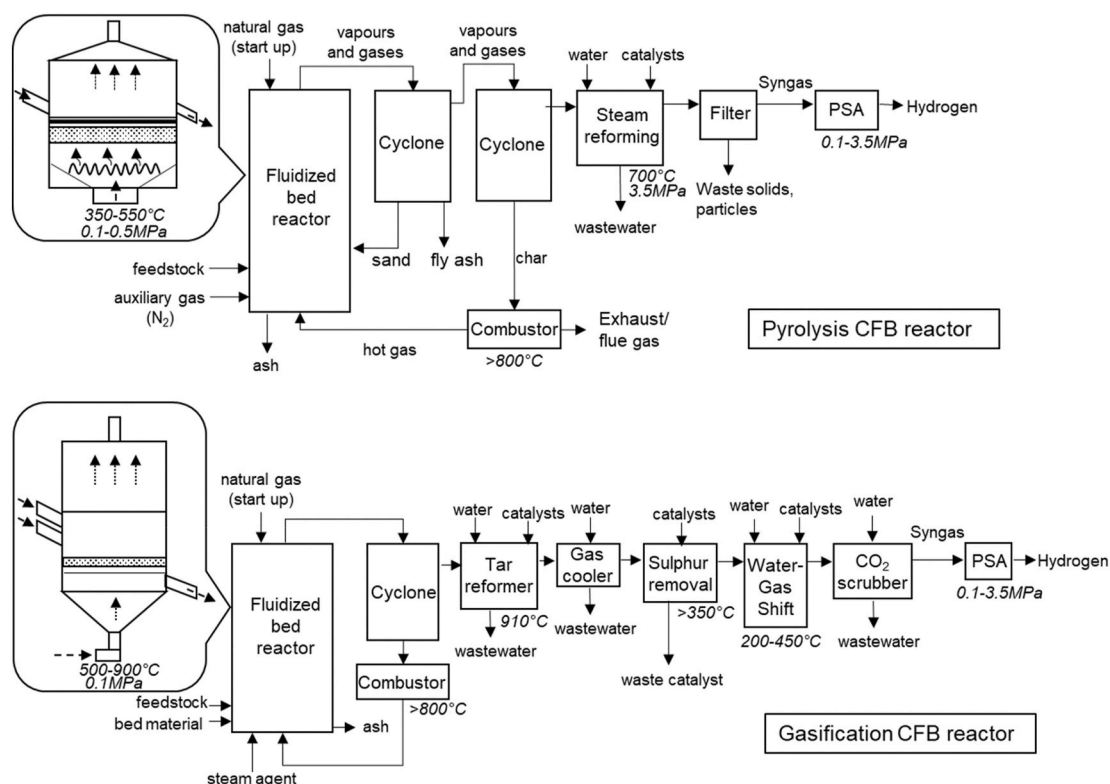


Fig. 1. Simplified flow diagrams of pyrolysis and gasification processes using circulating fluidised bed reactors (CFB). CFB reactors have high gas flow rates and are suitable for continuous hydrogen production.

3.1.1. Gasification

The gasification process (Fig. 1) is the conversion of solid material between the temperatures of 500 °C and 1200 °C in an oxygen-deficient environment at atmospheric pressure (0.1 MPa) [3]. Autothermal gasification uses partial oxidation of waste within the reactor, in the presence of an oxidant at an amount lower than that required for stoichiometric combustion, to provide the required heat for the reaction [39]. Part of the feedstock is combusted in exothermic reactions to provide heat to gasify the remaining products [40]. The reducing atmosphere of the process limits the emissions of furans and dioxins that often link the combustion of waste [41], while the oxygen-deficient condition reduces heat losses and increases energy recovery efficiency. Gasification of biomass for hydrogen production has a thermal efficiency of 35–50%, up to 52% for steam gasification [18]. While a study by Salkuyeh et al. [42] suggested that entrained flow gasification could have a thermal efficiency of 56%LHV and fluidised bed gasification could reach 45%LHV. Adaptations to reactor system technology and materials have improved the economics of the process and shown positive environmental effects [44].

Typical gasifying agents or oxidation media include air, oxygen-enriched air, pure oxygen or steam. Steam gasification endothermic and requires energy input to produce syngas with the quality comparable to the other oxidation agents [46]. Beyond the optimum temperature, the hydrogen yield decreases as the high temperature accelerates the reaction reducing the ability of cracking and reforming reactions [47]. The Steam to Biomass ratio (S/B) is important for steam gasification as the steam reforming reaction is the main reaction producing hydrogen [48]. While hydrogen increases initially after a certain S/B point, excess steam causes a decrease in hydrogen production and encourages tar formation. In general, larger fractions of steam added to the gasifier cause the S/B ratio to increase, CH₄ and CO to decrease, while CO₂ and H₂ to increase, whilst reducing tar formation [47]. Steam or oxygen gasification produces syngas of an HHV of 9.2–16.5 MJ/m³ at standard temperature (25 °C) and pressure (1atm) [49].

Air gasification has a heating value of 3.5–7.8 MJ/m³, which is low compared to steam and oxygen gasifying agents [43]. Using air as the gasifying agent increases the concentration of nitrogen (air contains ~79%) which dilutes the syngas concentration, increases the cost of separation and lowers the heating value [22]. However, air gasification is common and cheap due to air withdrawal from the atmosphere. Oxygen-enriched air is a compromise between heating value, abundance and cost. With oxygen content at levels between 21% and 50%, the reduction in nitrogen content increases the higher heating value of syngas [37]. Reduction in H₂ and CO yields also occur with increasing equivalence ratio (ER) (actual oxygen flow rate to stoichiometric oxygen flow rate), beyond the optimal range (towards the combustion zone ~1) reducing the heating value of syngas and producing a low-quality gas product. Beheshti et al. [50], showed that CO₂ increased (45.2 mol%) and H₂ increased (12.1 mol%) while CO decreased (12.9 mol%) as ER increased from 0.3 to 0.7 during the biomass gasification process. A decrease in tar content is also noted if gasification is conducted at a higher ER because of the increase in thermal cracking. The presence of volatile material and moisture, up to 15%, in the feedstock can increase the ER, while more than 15% causes temperatures to vary [22].

A variation from traditional gasification techniques is chemical looping gasification. This process uses solid metal oxides (Mn-, Cu- and Ni-) as the oxygen carrier to provide oxygen instead of pure oxygen in a looping system [43]. The reduced oxygen is regenerated and replenishes the oxygen which then returns to the gasification reactor [51]. Optimum operating conditions can produce a gas stream with high H₂ and CO contents whilst lowering CH₄ and CO₂, plus reduced tar generation. Udomsirichakorn and Salam [52] studied chemical looping gasification using calcium oxide (CaO), to control CO₂ capture and reduce tar formation. Chemical looping gasification can combat deactivation of CaO after carbonation which allows for continuous hydrogen production and improves the economics for hydrogen production.

Operating temperature has a large influence on the hydrogen yield of gasification [12], gasification kinetic rate and enhances endothermic

equilibrium reactions [46]. Rise in temperature towards the optimum increases the hydrogen yield due to an enhancement in reactions such as the Boudouard, water-gas shift and reforming reactions [47]. Sirirermux and Kerdsuwan [53] found that when the reaction temperature was raised from 700 °C to 800 °C then further to 900 °C, the energy output (kJ/kg of feedstock) increased by 1.5 and 2 times.

Catalysts are used in gasification to improve the hydrogen production rate, gasification efficiency, and promote tar cracking and steam reforming reactions [54]. Specifically, catalysts can lower the activation energy required for the reactions and improve the carbon conversion efficiency, leading to higher gas yields [55]. Traditional catalysts include mineral, nickel-based (Ni/Al₂O₃), alkali based, olivine and dolomite based [26]. Nickel-supported catalysts have been found to reduce the formation of tar, char (recalcitrant by-products) and CO₂ in gasification [55]. Due to the cost-effectiveness of dolomite and olivine, which are found in nature, they are more frequently used [22]. Biochar catalysts have been receiving increasing attention due to their low costs, porous nature and availability via gasification or pyrolysis of waste [57]. Ma et al. [59] used biochar as a catalyst to enhance the steam reforming of bio-oil to produce a maximum hydrogen yield of 89.13% and a concentration of 75.97%. However, catalysts can suffer from poisoning, plugging, loss of activity and deactivation including from carbon deposition [60]. Particularly, waste (e.g. sewage sludge and animal waste), may contain sulphur compounds, particulates and trace metals which will adversely affect the performance of a catalyst [43].

Co-gasification, multiple feedstocks with different properties, have been proposed to improve hydrogen production rates and reduce energy requirements [36]. The co-gasification process reduces the CO₂ emissions of gasification of separate feedstocks due to increased efficiency and reduced energy requirement [61]. You et al. [62] compared syngas production of the co-gasification of food waste and woodchips with sewage sludge and woodchips from research by Ong et al. [63]. Food waste and woodchips co-gasification produced 32.9% of syngas (regarding total gas composition) compared to 32.4% syngas from sewage sludge and woodchips co-gasification. While the volume of hydrogen produced was higher for sewage sludge at 16.8% compared to food waste at 16.5%, due to food waste having a higher energy potential and a lower ash content while sewage sludge contained higher moisture and ash contents. Ong et al. [63] also found that less than 33% of dried sewage sludge in the mixture was preferred due to the high ash content. Huang et al. [36] studied the co-gasification of wet sewage sludge and forestry waste with steam as the gasifying steam agent and found that gas yield decreased as the ratio of wet sewage sludge increased. The H₂ and CO concentrations were the highest when the ratio of wet sewage sludge was 50%. The maximum hydrogen yield (33.6 mol/kg) occurred at 1100 K and for spruce waste. Beyond this optimum temperature, increasing the ratio of sewage sludge in the mixture did not increase the yield of hydrogen yield due to the surplus moisture content and reduced organic matter.

Plasma gasification uses an electric arc to ionize an inert gas to form plasma which becomes the heat source of gasification for syngas production from waste such as unsorted MSW and melt inorganic material [64]. Typically, a plasma gasification reactor consists of one or more plasma arc torches (~13,000 °C) at atmospheric pressure and can produce a high temperature from 2000 to 5000 °C where the plasma interacts with the fuel [66]. Syngas reactions within the plasma gasifier are activated by the injection of steam. The residence times, flowrates of plasma gas, oxidant and steam streams plus reaction temperature affect the quality and mass of the gas produced [67]. The typical syngas composition is 15% H₂, 24% CO, 6% CO₂ and 55% N₂ with an increase in moisture and inorganic matter increasing the H₂ and decreasing CO content [68]. Increases in reaction temperature produce more net energy content with higher combustion enthalpy and reduce tar content in the syngas to less 10mg/Nm³ [67].

The plasma gasification method allows great flexibility in feeding rate, moisture content and composition of waste material, and thus is

suitable for mixed biomass feedstocks of variable particle size with minimal preparation [43]. Plasma gasification produces very small volumes of vitrified slag, and emissions such as particulate matter, mercury and metals requiring pollution control devices (e.g., scrubbers for syngas cleaning, cyclones and water quenchers) [69]. Hence, the emissions of plasma gasification are much less when compared to other types of gasification [65]. Ramos et al. [69] conducted a life cycle analysis on two-stage plasma gasification for MSW and showed that it was environmentally friendly and sustainable. However, plasma gasification has not been proven for large-scale industrial applications yet [70].

After gasification syngas requires cleaning, reforming and gas shift reaction processes, and separation processes to generate high purity hydrogen (99.9%) [49]. Warm or hot gas cleaning techniques can improve thermal efficiency if integrated into combined cycle gasification systems [71]. Pressure swing adsorption (PSA) is a conventional method of hydrogen separation and it utilises an adsorbent bed to capture impurities in the syngas at high pressure which are subsequently released as the pressure is reduced [18]. The PSA method requires a minimum of 70 mol% hydrogen in the input gas stream [27] efficiency as high as 99.99% [73]. PSA systems can differ according to adsorption size, velocity, regeneration and adsorbent material [72].

Membrane technologies can also be used to adjust the gas composition in syngas [39]. The partial pressure of the hydrogen feed streams forces the permeation and balances with the product stream. Most membrane materials are expensive and susceptible to contamination in the syngas even after cleaning-up processing [72]. Using zeolitic frameworks, porous materials with metal nodes linked by imidazole ligands, can isolate selected gases in syngas, leading to increases in the ratios of H₂/CO and H₂/CO₂ [74]. The thermal stability of the zeolitic framework at high temperatures (above 230 °C) is poor though Yang and Chung [75] have shown that zeolitic imidazolate frameworks-8 nano polymer as a composite material can have high thermal and separation stability and H₂/CO₂ selectivity. A sweep gas (nitrogen) can be used on the other side of the membrane to lower the partial pressure and encourage more hydrogen to pass through [18]. Other separation methods include temperature swing adsorption and electrical swing adsorption, and cryogenic processes (requires extremely low temperatures). However, these methods are still in the experimental stage or expensive to run and are currently not commercially viable [71].

3.1.2. Pyrolysis

Pyrolysis is a thermochemical process (Fig. 1) that can convert carbonaceous materials into value-added products such as bio-oil, biochar, and product gas at temperatures of 350–550 °C, and pressures of 0.1–0.5 MPa in the absence of oxygen [9]. The lack of air mitigates the formation of dioxins [73]. Pyrolysis can be performed before gasification to generate biochar for gasification to improve the energy content of gasification feedstock. The hydrogen-containing product gas can be transformed via steam reforming and water gas shift reaction to produce more hydrogen [76]. The gas yield is dependent on the feedstock type, temperature, heating rate, catalyst, and residence time of the process [77]. The use of catalysts based on inorganic salts such as chloride, carbonates and chromates can speed up the pyrolysis process [73].

Table 2 summarises the hydrogen production (percentage or yield) of gasification and pyrolysis under a variety of process conditions from various studies. Hydrogen concentrations ranged from 29.5% to 82.01% which are normally associated with biochar or bio-oil. Temperature does not appear to be related to the hydrogen production volume from a specific feedstock, e.g., higher temperatures do not consistently match higher hydrogen concentrations. MSW corresponds to lower hydrogen concentrations and yields compared with other types of feedstocks.

3.2. Biochemical processes

Biochemical processes for hydrogen production (biohydrogen)

Table 2

Summary of hydrogen production of gasification and pyrolysis under different process conditions. The hydrogen yield or concentration suggest the process efficiency. (MSW = municipal solid waste).

Author	Feedstock	Main Reaction	Agent/Catalyst	Reactor	Op Temp °C	% H ₂ conc	H ₂ yield
Chen et al. [31]	Bio-oil/biochar slurry (from pyrolysis of pine sawdust)	Catalytic steam gasification	La _{0.8} Ce _{0.2} FeO ₃ perovskite type catalysts	Fixed bed	800 (pyrolysis at 550)	82.01% with C conversion 65.57%	–
Yao. J et al. [78]	90 wt% bio-oil and 10 wt% biochar	Steam gasification	LaCo _{0.9} Cu _{0.1} O ₃ perovskite type catalysts	Fixed bed	800	75.33% with C conversion at 80.42%	–
Ma et al. [59]	Bio-oil	Catalytic steam reforming	Biochar catalyst	Fixed bed	900	75.97% conc	89.13%
Zeng et al. [79]	Biochar	Steam gasification	Steam	Fixed bed	800	74 vol% conc	0.0714 kg/kg
Chen et al. [58]	Biochar from rice husk for bio-oil	Steam reforming of bio-oil	Ni/BC4 catalyst (biochar activated)	Fixed bed	700	71.20%	–
Chutichai et al. [47]	Saw-dust	Water-gas shift reactor and oxidation	–	Fluidised bed gasifier	600–650	–	33 mol.%
Al-Rahbi and Williams [60]	Sawdust wood pellets	Steam gasification	Tyre char	Two stage fixed bed	900	56 vol%	39.20 mmol g ⁻¹
			Acid treated tyre pyrolysis	Two stage fixed bed	900	–	30.4 mmol g ⁻¹
Xin et al. [80]	Cattle manure	Two step gasification	–	Fixed bed	850 (500 pyrolysis)	57.58% conc	0.93 m ³ /kg
Yao. D et al. [81]	Wheat straw (biochar)	Two step steam gasification	Ni/cotton char catalyst	Two stage fixed bed	800 (550 pyrolysis)	64.02 vol% conc	92.08 mg g ⁻¹ biomass, gas yield about 90 wt %
Li et al. [82]	Corn stalk	Steam gasification	CaO	Fluidised bed gasifier	650	61.23 vol%	493.91 mL/g biomass
Chang et al. [40]	Commercial α-cellulose and agricultural waste	Gasification	–	Fluidised bed gasifier	1000	29.50%	–
Hu et al. [34]	MSW	In situ steam gasification	–	Fixed bed	750	49.42 vol% conc	277.67 mL/g MSW
Sirirermux et al. [53]	MSW	Steam gasification	Steam agent	Drop tube fixed bed	800	–	34.34 gH ₂ /kg MSW
Williams and Wu [76]	Plastic (polypropylene)	Pyrolysis and catalytic gasification	Ni–Mg–Al catalyst	Screw kiln and Fixed bed gasifier	500 (pyrolysis) and 900 (gasif)	41.65%	17.87 g/100 g plastic
Klaas et al. [83]	Hemp seeds	Pyrolysis	Steam reform-ing/no catalyst	Fluidised bed pyrolysis reactor	700	–	2 wt % origin biomass convert to H ₂
Wickramasinghe and Narayana [84]	MSW	Torrefaction	Nitrogen agent	Batch type fixed bed torrefaction	350	–	–

Table 3

Summary of hydrogen production of biochemical methods. Results are represented by hydrogen yield. VSS = volatile suspended solids, VS = volatile solids, FW = food waste.

Author	Main Reaction	Feedstock/Substrate	Bacteria/Inoculum	Reaction Parameters	H ₂ yield
Eker and Sarp [29]	Anaerobic dark fermentation	Glucose from acid hydrolysed wastepaper	cultivated heat treated anaerobic sludge	Fermentation medium with C/N/P ratio 100/2/0.5. Initial pH6.8, ORP -350 ± 25 mV	140 mL H ₂ /g total sugar at 3.84 g/L total sugar conc
Liu et al. [94]	Two stage mesophilic fermentation	Activated sludge and food waste	Hydrogen producing bacteria	Pretreated with heat (cultured, 100 °C for 30 min s) VS/TS 51.8%. Initial pH 5.5, temperature 37 °C	14.0kJ/g-VS at food waste of 100%
Cheng et al. [95]	Combined dark- and photo-fermentation	Alanine and Serine amino acids from waste biomass	Clostridium butyricum and Rhodospseudo-monas palustris	Amino acid + HPB inoculum + deionised water mixture, initial pH6±0.1. N ₂ purge, temperature at 35 ± 0.1 °C	418.6 and 270.2 mL/g
Wang et al. [86]	Effect of biochar on fermentation	Dewatered activated sludge and food waste	H ₂ production inoculum form biogas plant of local brewery	Feedstock air dried, then pyrolyzed. Substrate, inocula and water initial pH5.5. Different biochar added. N ₂ purge, temperature at 35 °C, stirred at 150 rpm.	47.2–83.6 mL/day
Wang et al. [88]	Adding calcium peroxide to dark fermentation	Waste activated sludge	Hydrolytic microbes	Sludge had pH6.9 ± 0.1, TSS of 14.5 ± 0.3 g/L, VSS of 11.8 ± 0.2 g/L. CaO ₂ added to mixture. N ₂ purge. Air-bath shaker at temperature 35 ± 1 °C, 200 rpm.	10.55 mL/g VSS
Pu et al. [87]	Varying substrate concentrations on H ₂ production during fermentation	Heat treated and fresh food waste	Inoculum seed sludge	Fresh/food waste heat treated then added to inoculum sludge with water. Initial pH6.5, N ₂ purge. Shaker at 100 rpm at temperature of 37 °C	75.3mL/g-VS at 15 g-VS/L heat treated FW
Sharma and Melkania [92]	Anaerobic fermentation-add biochar to improve hydrogen production	Organic MSW	Enterobacter aerogenes and E. coli	Feedstock shredded and filtered. N ₂ purged. Inoculum, biochar and medium at initial pH5.5 and temperature of 37 °C	96.63 ± 2.8 ml H ₂ /gCarbon _{initial}

utilise microorganisms to decompose waste products containing organic material, biomass or wastewater as substrates to produce hydrogen [85]. Typical processes include photo or dark fermentation or a hybrid system [26]. Most biochemical processes are less energy-intensive than thermochemical processes as they occur at ambient temperatures and pressures, however they have low hydrogen yields and low reaction rates. Table 3 summarises the hydrogen yield data and associated waste feedstock from various biochemical studies and current research. The range of microorganisms studied suggests waste with different characteristics can be matched with a specific bacteria/inoculum leading to optimum production rates in generally otherwise low-efficiency processes.

3.2.1. Dark fermentation

Dark fermentation (with anaerobic bacteria on carbohydrate-rich substrates in anoxic and dark conditions) is characterised by its ability to treat a variety of feedstocks (e.g. food waste [87], paper waste [29], waste activated sludge [88]) low energy demand, and the possibility to integrate with other processes such as methane production processes [29]. Fermentation breaks down carbohydrate-rich and biodegradable biomass to form organic acids, alcohols, acetone and then hydrogen and CO₂ [86]. Glucose (via glycolysis) is the preferred substrate of the process yet while it is expensive it is possible to generate glucose from agricultural waste [73]. Higher substrate concentrations will lead to lower hydrogen yields, because of the thermodynamic limitations imposed by microbial fermentation [89]. Other influential factors of hydrogen yield include mode and reaction conditions such as microorganism types. The metabolism of microorganisms is further affected by temperature, pH (ideal pH5 - 6), the partial pressure of hydrogen, pure or mixed bacterial cultures, method of preparation, hydraulic retention time for continuous processes [73], and composition of the medium [2]. Removing the hydrogen produced prevents a significant pressure increase which negatively affects production [18] and is important for continual production [2]. As the partial pressure of hydrogen increases hydrogen synthesis decreases and other products such as lactic acid and ethanol increase in concentration.

3.2.2. Photo-fermentation

Photo-fermentation produces hydrogen and CO₂ by using photosynthetic bacteria in anaerobic conditions at an optimum temperature of 30–35 °C with pH7 [73]. The process occurs for some microorganisms in nitrogen-deficient conditions using organic acids and sunlight. Some photosynthetic bacteria contain nitrogenase and can convert organic acids such as acetic, lactic, butyric, into hydrogen and CO₂. Rai and Singh [85], discussed a type of photo-heterotrophic bacteria called purple non sulphur bacteria which utilised a range of light intensity (approx. 3000–15000 lux or lumens/m²) and led to a high substrate conversion efficiency for waste and organic-rich wastewaters. The use of wastewater as feedstock requires pre-processing due to the colour limiting light penetration, possible contaminants and toxic compounds in wastewater [90]. Low light conversion efficiency affects production rate and yield along with microbial strain and carbon source.

Hybrid systems combining non-photosynthetic or anaerobic and photosynthetic bacteria have been proposed to reduce light requirements and increase hydrogen yields. Rai and Singh [85] conducted sequential dark/photo-fermentation with anaerobic bacteria placed under dark conditions producing hydrogen and organic acids, which became sources for photosynthetic bacteria to produce extra hydrogen. The hydrogen yield increases as temperature rises for a stable pH4.5-6.5 for fermentation and above pH7 for photosynthetic bacteria [18]. Lin et al. [91] looked at the increase of bioenergy production efficiency from two-stage fermentation processing of wastewater. They suggested that combined dark and photo fermentation had a theoretical hydrogen yield of 12 mol/mole glucose. Wang et al. [86] found that adding biochar could improve the maximum H₂ production rate and shortened lag time for the fermentation process. Their analysis suggested that biochar had a

strong pH buffering capacity and stabilising effect during hydrogen production. Sharma and Melkania [92] found adding biochar to the anaerobic fermentation process of organic MSW using a co-culture, shortened the lag by 4.4 ± 0.5 h and increased the hydrogen production. Sun et al. [93] added biochar and metal co-factor nanoparticle Ni⁰ to dark fermentation and found it to increase hydrogen yield. In part due to increasing the pH buffer stability as well as enhanced electron/photon transfer plus the facilitation of biofilm formation during the initial fermentation.

Current technology advancements are focussing on efficiency improvements, increasing energy recovery, reducing additional energy input requirements, waste heterogeneity issues and upscaling. Technology growth is limited by uptake and economic cost, though hydrogen production projects in transportation industry showed that some governments and investors are willing to be early adopters, as addressed in the following sections.

4. Applications and bottlenecks

Highlighting applications for hydrogen goes some way to justifying the cost and development of conversion technologies. Identification of technology and social bottlenecks allows research and policy to focus on specific targets.

4.1. Application of hydrogen

The status and development of WtH technology are occurring in conjunction with an increase in the popularity of hydrogen as a fuel. The transport sector is the second-largest consumer of hydrogen [96]. Transition and integration to a hydrogen society are in part driven by the ambition to reduce fossil fuel use and the growing concern for climate change. Behling et al. [97], Itaoka et al. [98] and Matsuda and Kubota [99] detailed the government strategy for development into a hydrogen society which includes developing hydrogen fuel cells and setting out a “Hydrogen Highway”, a plan for integration of hydrogen technologies. Sørensen et al. [100] used management programmes and decentralisation of energy production as markers to determine the transition scenario to a hydrogen society.

The hydrogen technology development is driven by demands for end-use applications such as fuel cells for vehicles. The number of electric vehicle sales doubled to over 1.2 million in 2018 [1]. Hydrogen fuel cell vehicles have the potential to be an alternative to electric vehicles as they provide onboard electricity generation with the ability to fill at stations similar in design to petrol stations. Disadvantages of electrical vehicles over hydrogen vehicles include the short to limited driving range, relatively long recharging time (hours), limited recycling options for used batteries [2].

A fuel cell is an electrochemical device that converts the chemical energy of gas such as hydrogen or a solid such as coal into electrical energy [101]. A fuel cell contains an anode where the hydrogen gas splits into ions (H⁺) and negatively charged electrons (e⁻). The ions pass through an electrolyte with the electrons being forced around the outer circuit towards the cathode forming an electric current [24]. The ions combine with oxygen from air forming water as a green by-product with no other emissions. Major types of fuel cells include Proton Exchange Membrane Fuel Cell (PEMFC), alkaline, Solid oxide fuel cell (SOFC) featured by different power densities, hydrogen production rates, and specific energy consumption (kWh) [20]. They can be compact in size and portable, therefore desirable for vehicles as well as stationary power generation units [24].

Fuel cell projects currently seen in transport sectors are demonstration projects in cities such as Aberdeen, Oslo, Hamburg and Zhengzhou [102]. Polymer electrolyte membrane fuel cells (PEMFC) are a popular fuel cell technology that is mature for commercial development [103]. The fuel cells operate at lower temperatures (80–100 °C) and pressures, with a high-power density, low maintenance requirement and being

light weight [104]. However, PEMFC requires pure hydrogen due to its low temperatures (approx. 70 °C) and the use of precious metal-based electrodes [47]. This has implications for potential feedstocks due to impurities present within waste derived hydrogen.

Adding value and finding alternative usage for the produced hydrogen, other than transport applications, will improve the appeal of a transition to the hydrogen economy [105]. Applications include heat for buildings and joining the energy grid by adding a hydrogen fraction to the natural gas network [106]. If the fraction is delivered by renewable sources the gas network would gain green credentials and step further towards reducing the carbon footprint of energy.

4.2. System development

The infrastructure of a WtH system comprises plant/reactor sites, storage facilities, delivery and transport options, refuelling stations, conversion and end-use applications (Fig. 2). Additional considerations are those related to safety and technical issues of using hydrogen gas with design factors including separation distances and a low fire-resistance rating of onboard storage [107].

Ideal locations for the development of biochemical systems are those close to sources of raw material to limit cost and transportation. This depends on the identified optimum feedstock or available mix for the specific operational requirements or composition of syngas expected. The criteria for selecting raw material is availability and source location, as well as cost, carbon content and biodegradability [73]. An ongoing issue for WtH gasification plants, causing both technical and economic problems, is the heterogeneous nature of waste feedstock especially for household waste [18]. The cost of materials transport and energy requirements will be affected by the density, form and moisture content of waste, which needs to be considered in the plant planning stage.

4.3. Bottlenecks

Bottlenecks that limit or slow development exist throughout the hydrogen production system, from waste supply to conversion technologies, storage, and demand from customers. All parts of the process require support and technological innovation to increase and encourage growth in the hydrogen, waste recovery and management industries.

4.3.1. Competition

The petroleum industry continues to be profitable with reliable services and supply due to mature technology and structured organisations with decades of experience. This is despite cyclical upturns and downturns in supply and demand. To reach climate targets the long-term outlook for the role of fossil fuels will change, nevertheless short-term usage looks to remain. The goals for the hydrogen industry are overcoming the bottlenecks and being competitive as an energy carrier. WtH technologies need to be cost-competitive and offer highly technical and operational reliability along with environmental sustainability [37]. Uptake of the new technologies rely on policymakers encouraging and

investing in the hydrogen society. Meanwhile, support from all divisions of the energy industry and public is vital for hydrogen to become a contributor towards the low carbon and zero-emissions future [108]. However, a lack of information for the broader public on zero-carbon energy schemes can affect the uptake and the involvement from the general public [109].

4.3.2. Waste pre-treatment

Pre-treatment processes such as drying, and contaminant removal are essential to improve the energy density and for control over the physical and chemical properties of the waste before further processing. They require energy, time, space, transport, separation facilities, equipment and chemicals [33]. Solid waste management is associated with potential pollution for air, soil and water pollution [41]. Consequently, all practices involving waste require monitoring throughout the process chain to avoid mishandling and pollution. High-end waste management technologies such as the ones based on GIS, radio frequency identification, ultrasonic sensors and international system for mobile or general radio packet service, have been proposed to enhance waste collection and preparation [9].

4.3.3. Storage (waste and hydrogen)

The solid waste management chain consists of waste generation, collection, segregation, storage, transport to treatment pathways and final utilisation [9]. Storage for waste depends on quantity and capacity of processing plants, which is generally affected by continuous or intermittent waste supply. Movement between collection, processing and feedstock conversion sites relies on efficient communication and management with accurate forecasting of volumes using computational systems and models to avoid congestion and backlog [9].

Hydrogen storage options are currently limited, with the common choice being gas compression at 35–70 MPa and 300 K (ambient temperature) requiring 5–20% LHV energy requirement [23]. Liquid hydrogen as the other option is stored in cryogenic tanks requiring compression and cooling [18]. Solid-state hydrogen storage has been explored which involves physical storage of molecular or di-hydrogen in nano-porous materials such as activated carbons [110]. Solid-state storage based on metal hydrides has shown promise according to Abe et al. [111], who state they are secure, compact, capable of holding large quantities of hydrogen and are repeatedly reversible. Chemical storage options include complex hydrides such as magnesium borohydride Mg(Bh₄)₂ and sodium borohydride was found to exhibit the highest gravimetric densities (1.074 specific gravity) for hydrogen storage [107]. While fuel cells containing borohydride have potential as hydrogen storage [112]. Research challenges for chemical storage include capacity improvements, kinetics of uptake and release of hydrogen considering that the kinetics for bulk materials often require high temperatures (approx. ~400 K) and are slow [107]. Current hydrogen storage alloys have a low capacity typically less than 2% mass of H₂, therefore advancements in material technology (alloying, catalysis and milling) and in lowering temperatures and increasing rates of dehydration will be

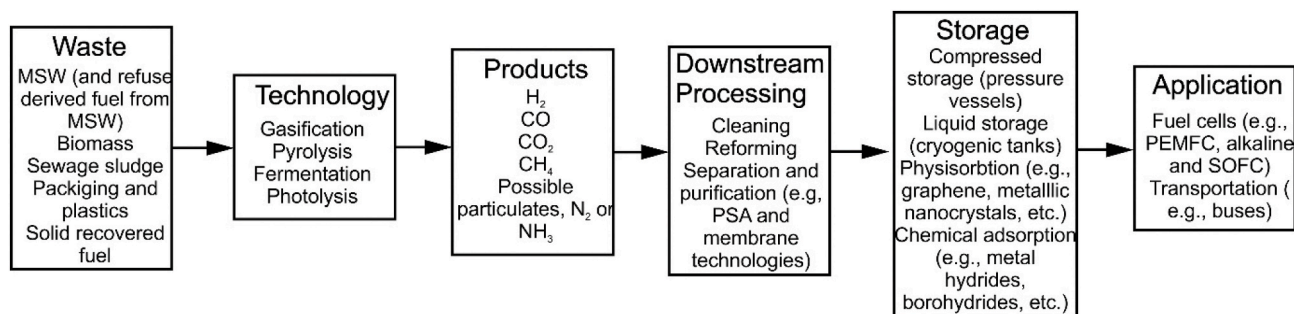


Fig. 2. The schematic diagram of WtH process flow with the main stages and products. The arrows indicate flow direction or transport between the different stages.

required [113].

Storage options impact the scale of hydrogen production when considering future upscaling and large industrial applications [21]. Lai et al. [113] suggested that storage was one of the main obstacles to widespread hydrogen use requiring better storage material design for transport and distribution purposes. Stockfield et al. [107], suggested that improved practicality of storage options is critical to increase implementation capacity and improve operating conditions.

4.3.4. Transportation of hydrogen

The hydrogen distribution network is currently underdeveloped and immature. An increasing number of countries and regions (Europe and Asia) are investing in the development of hydrogen infrastructure, market and vehicles, escalating the demand for hydrogen. The main transportation and delivery methods include road (trucks for liquid and gas hydrogen) and railway for storage containers and vessels. Due to volume limitations and low-density, the cost of products including gas is relatively high [114]. The conversion of natural gas pipelines to hydrogen pipelines adds the potential for additional delivery locations and areas. Pipelines for transporting compressed hydrogen can have dense networks in some areas such as the ones of the chemical industry [115], while may be limited in other industrial areas creating an inconsistent network requiring extra maintenance effort [116]. Improvements in technology with the aim to improve transportation efficiency would be required to reduce H₂ pipeline losses which could be as high as 20% [18].

4.3.5. Processing technologies

The technical bottlenecks of the thermochemical processes generally include low conversion efficiencies, low hydrogen yields and the generation of unwanted by-products such as tar in the product gas incurring additional cleaning effort and cost [43]. Specifically, for gasification, it is critical to decrease the cost of syngas cleaning, to achieve higher energy efficiencies, and have great flexibility in meeting defined gas specifications [37]. These issues are summarised in Table 4, highlighting the many bottlenecks attached to waste processing technologies of interest.

Other issues with the thermochemical processing of waste involve the presence of alkali and earth alkali metals (mainly potassium) [117] which become components of ash that require removal and filtering to prevent fouling and agglomeration of related processing equipment [118]. For MSW gasification, as detailed below, the content of tar should be no more than 1 mg/m³ in the product gas or syngas [43]. The characteristics and amount of tar formed depend on fuel type, operating conditions and secondary gas-phase reactions (e.g., chain radical reactions, reactions of aromatics species, and molecular dehydration reactions) [70]. The condensation temperature of tar is between 200 and 600 °C, and within this range damage to equipment and operation issues can occur including gas passage clogging, deactivation of sulphur removal systems, and damage to gas engines [119]. If the tar is transferred to wastewater i.e. through gas tar condensation, droplet filtration, or gas/liquid separation, extra treatment and disposal is required leading to energy loss [70]. To reduce alkali tar and ash fouling, specific bed materials can be used such as alkali feldspar, olivine, and low iron bauxite [117].

For biochemical processes, bottlenecks exist regarding efficiencies which hinder larger uptake of the technology [120]. Generally, biochemical processes are featured by low efficiency, low yield and slow rate of reactions the processes [115]. Additionally, species of microbes and hydrogen-producing bacteria require certain light intensities and specific conditions for photo-fermentation [95]. These variations may cause yields to become unpredictable and impact the hydrogen chain and source reliability, therefore further affecting growth and uptake of hydrogen as a fuel.

Research focus for dark fermentation is on finding, isolating and improving strains of bacteria for the conversion of lignocellulosic

Table 4

Summary of the main comparisons between the processing technologies mentioned and the influences for these parameters.

Processing Technology	Main issues	Influences
Steam Gasification	High cost of pre-treatment High cost of syngas cleaning Low thermal efficiency	Drying, grinding and shredding to improve energy density of waste Tar/unwanted compounds cause fouling of equipment without cleaning. Variable waste composition and character
Air/Oxygen Gasification	High cost of pre-treatment Low thermal efficiency High cost of syngas cleaning Low thermal efficiency	Drying, grinding and shredding to improve energy density of waste High nitrogen content of air agent reduces energy content of syngas Tar/unwanted compounds cause fouling of equipment without cleaning. Variable waste composition and character
Pyrolysis	High cost of syngas cleaning Low thermal efficiency	Tar/unwanted compounds cause fouling of equipment without cleaning. Variable waste composition and character
Dark fermentation	Low conversion efficiency	Microorganisms can suffer from inhibition from increased hydrogen partial pressure, presence of other organisms causing unproductive conditions
	Slow reaction times	Rely on the metabolism of microorganisms which are slower than thermochemical methods
	Low hydrogen yield	Microorganisms limited by waste (substrates) they can consume
Photo fermentation	Unpredictable yields	Microorganisms require specific conditions (light, partial pressure), changes in will affect yield
	Slow reaction times	Current microorganisms have slow work rates
	Low hydrogen yield	Microorganisms limited by waste (substrates) they can consume

material to hydrogen. One option is genetic engineering of microorganisms to improve metabolism and the yield of hydrogen [2]. Ongoing research into using lignocellulosic for the production of biofuels such as bio-oil and bio-ethanol exists though efficient technology for the industrial production of hydrogen is still unavailable [118].

4.3.6. Fuel cells

Issues can arise with using waste as a source of hydrogen as hydrogen contamination can occur and requires significant cleaning and purification efforts. For example, the syngas generated from the gasification process a significant amount of CO which can deteriorate PEMFC performance [47]. A purification process is needed to decrease the CO content (10 ppm or less) to avoid catalyst poisoning. Chutichai et al. [47] found that the CO concentration was reduced by flowing the product gas through high and low water gas shift reactors and an oxidation reactor, leading to 33 mol% H₂ in the final product gas with a decreased CO concentration (<10 ppm) with an overall process efficiency of 57%.

SOFCS are better at handling lower purity hydrogen so are desirable for the hydrogen produced from waste [18]. They can also run on natural gas, biogas, shale gas and coal gas, thereby increasing the flexibility and resilience of a hydrogen system [24]. However, they suffer from the problems of high costs, low durability, difficulty in optimisation of interfaces with other technology, and limited performance and sustainability of materials [17]. SOFCs require rare earth elements and use a large volume of lanthanides (metallic elements) therefore the infrastructure around SOFC manufacture may be limited with doubt over access to alternative materials that are sustainable and cheaper [107].

5. Environmental impacts and economic feasibility

For waste-to-hydrogen technologies to be included in the global energy mix both environmental concerns and economic viability require research supported indications on the benefits.

5.1. Environmental impacts

The WtH concept intends to reduce the volume of waste sent to landfill and the associated environmental pollution. It was estimated that an EfW plant processing 1 Mt per year for 30 years required less than 100,000 m² of land compared to 300,000 m² for landfilling of 30 Mt of MSW in the USA [121]. WtH technologies can form a complementary partnership with recycling activities, as they encourage additional sorting and pre-processing of metals, glass, and inorganics. Fig. 3 illustrates a potential waste management system with thermochemical and biological processes included alongside landfill and incineration practices.

The process of using waste for resource recovery whether it be hydrogen or electricity produces energy as a valuable product to displace fossil fuels [122], leading to an additional environmental benefit-GHG emission reduction. The global warming potential (GWP) indicates the amount of greenhouse gas (kg CO₂ equivalent) emitted throughout the life cycle of a technology, process or system [123]. The GWP will be adversely affected by the lower efficiency of hydrogen production technologies. Achieving and maintaining a low GWP would help boost WtH technologies while development and infrastructure becomes more competitive. The GWP of hydrogen production can be reduced if renewable resources are used at all stages and all products created are utilised [116]. Pandey et al. [124] stated that hydrogen production from biomass feedstock has the potential to reduce CO₂ emissions when compared to SMR. Valente et al. [125], study on eco-efficiency found hydrogen from biomass gasification was 5–38 times higher than SMR produced hydrogen when using GWP as an indicator.

Fujii et al. [126] conducted an LCA proposing the use of waste in energy production methods including upgrading of WtE to improve exergy efficiency as well having the potential to increase CO₂ emission reduction from 0.28 kg-CO₂/kg waste to 0.67 kg-CO₂/kg. They also discuss the formation of a hybrid industry to diversify current fossil fuel resources with recycle and renewables to lower carbon emissions. Currently SMR from natural gas followed by coal gasification are the most cost-effective forms of syngas production [18], commercial hydrogen plants produce emissions of 9–11 kg CO₂/kg H₂ in typical natural gas plants [42]. The use of fossil fuels to produce hydrogen without the use of CCS is therefore carbon-intensive. Carbon capture and storage (CCS) is an expensive and energy-intensive way to deal with CO₂ emissions from fossil fuel-derived hydrogen. Carbon taxes will directly influence the hydrogen production cost [18].

5.2. Economic feasibility

As the WtH industry is expanding, the initial cost of hydrogen production for transportation remains high due to limited infrastructure and options for the manufacture of components, and underdeveloped supply chain. The early stage of WtH development depends on pioneer firms and innovative projects while costs are high with relatively low efficiency. Ajanovic and Haas [108] stated the main barrier to large scale uptake of hydrogen is that it is currently is not economically feasible. Salkuyeh et al. [42] showed that the cost needed for hydrogen production from biomass gasification was competitive with SMR under the conditions of a minimum price of the biomass of \$100/tonne, \$115/tonne CO₂-eq., or minimum natural gas price of \$5/GJ. The economic evaluation from Yao et al. [127] displayed an optimal equivalence ratio (ER) for maximising the economic benefits of gasification. Their study demonstrated that as ER increased from 0.1 to 0.6, the HHV decreased from 6.15 to 3.60MJ/Nm³. Their model predicted a maximum economic benefit at \$0.11/kg biomass feedstock and an optimum ER of 0.25.

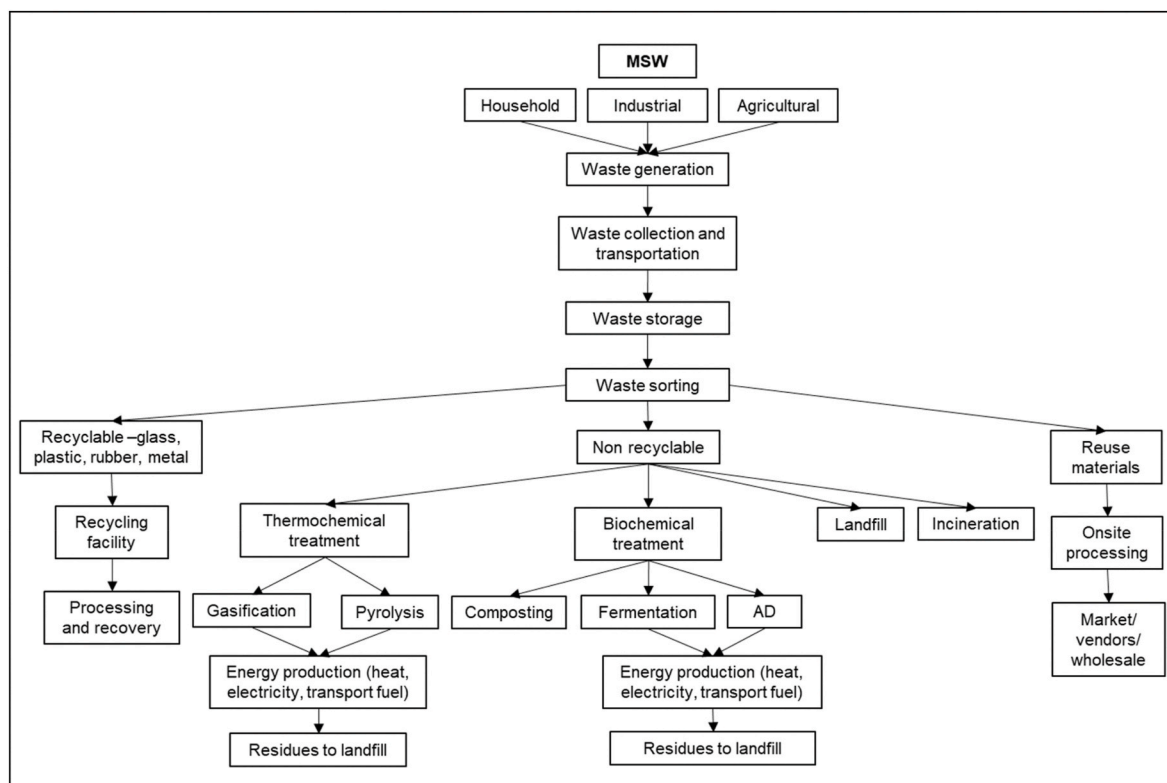


Fig. 3. Schematic diagram of waste management flow for municipal solid waste (MSW).

The demand for hydrogen in transportation is connected to the demand for fuel cells and accordingly the cost of fuel cells. Hydrogen cost reduction is essential for developing hydrogen-based transportation [108]. Waste can be a relatively expensive resource due to its physical properties of low bulk density, expensive to gather, store, process, handle and transport [77]. Transport of MSW from households to processing plants for large towns and cities can add significant costs depending on the delivery systems available especially if large distances are involved [114]. The sale of the hydrogen to transport companies will create revenue for the generation process, the amount will depend on the market value of the hydrogen gas.

Capital and operation costs of a gasification plant vary depending on; the size and processing requirements of feedstock, the end use of the syngas and consequently the complexity of the plant and the individual components. Capital costs include; storage, transportation, gasification system, gas cleaning system and energy generation system. The externalities include those for climate change and human health, emission and pollutant costs [125]. Other operation and maintenance costs include feedstock collection and handling, pre-treatment, transportation, grower payment, handling operating costs-chemicals, bed materials, catalysts, and processing of the syngas. Han et al. [128] conducted a techno-economic evaluation on fermentative hydrogen production on food waste and included factors such as total capital cost, annual production cost, annual profitability and sensitivity analysis. They found the process was feasible with a return on investment of 26.75%. Valente et al. [125], performed a cost assessment comparing the biomass gasification process against SMR found levelized cost of hydrogen from biomass gasification cost 3.59 € compared to 2.17€ for SMR. This covers the economic lifetime of the plant and the amount of energy produced from the plant.

6. Recommendations

After reviewing the data and research studies available the following sections contain suggestions for improvements that would advance waste-to-hydrogen development.

6.1. Efficiency improvements

It is critical to increase the efficiency of WtH technologies so that they can become cost-competitive to fossil fuels. The range of thermal efficiencies for the different WtH technologies with associated feedstock is listed in Table 5. Current efficiency ranges for the different techniques show the variability in what is being achieved and the potential for improvements to be made. Efficiency improvements can be made throughout the waste generation to hydrogen production steps, within the fuel and equipment supply chain (Fig. 2).

Types of waste feedstock with specific chemical composition as well as size, shape and moisture content will affect the reachable reactor efficiencies and therefore the hydrogen yield estimated. Reducing

Table 5

The efficiencies for the main hydrogen technology sourced from literature. Efficiency percentages, either thermal or process, broadly represent the ranges possible for the different technologies designed to highlight their variation.

Technology	Feedstock	Efficiency	Reference
Gasification	Biomass	35–50% thermal	Nikolaidis and Poullikkas [18]
Pyrolysis	Biomass	35–50% thermal	Nikolaidis and Poullikkas [18]
Plasma Gasification	MSW	43.3% process	Janaireh et al. [65]
Fermentation	Wastewaters	<30% process	Baeyens et al. [130]
SMR	Methane	70–75% thermal	Ju et al. [4]
Water Electrolysis	Water	75–80% thermal	Chisholm and Cronin [20]

uncertainty of the characteristics of the feedstock will improve the outlook for plants and production projects. Methods to increase energy density of biomass and improve the efficiency of a thermochemical process include pyrolysis and torrefaction which convert waste into carbon-rich biochar with a higher carbon ratio compared to oxygen and energy density [127].

Torrefaction is a mild pyrolysis process (heating at 200–300 °C in an inert atmosphere). Chen et al. [31] showed that torrefied biomass can achieve a heating value of 16–29 MJ/kg compared to 15–25 MJ/kg of raw unprocessed biomass. Torrefied biomass can retain 96% of the chemical energy and therefore improves the thermal efficiency for gasification [45]. Oxidative torrefaction, wet torrefaction, and steam torrefaction are alternatives to conventional dry torrefaction. The main product is biochar or carbon-rich torrefied material which via gasification can produce high-quality syngas through a higher efficiency process [84]. The change in biomass properties after torrefaction includes lower atomic ratios of O:C (0.4-0.4 to 0.1-0.7) and H:C (1.2-2 to 0.7-1.6) due to decrease in moisture, oxygenated and light volatiles, while the carbon remains [31]. A decrease in the volatile matter, lower moisture content, higher water resistivity, improved grindability, reactivity, and changes in density and hardness are also seen after torrefaction. To be feasible the extra energy required for both the torrefaction and the subsequent pellet making process are required to balance with the savings made though reduced transport, storage costs and overall efficiency enhancement.

Operational improvements and cutting energy requirements of the system aims to keep the energy input for hydrogen production below the energy available for secondary uses [19]. Research into different storage options will improve operational efficiency and therefore supply chain effectiveness allowing a more predictable and workable system. H₂FC (UK hydrogen community) has a hydrogen supply chain (HSC) model that can design and optimise the whole system. The model includes the spatial element and multiple timescales and can be used to map and analyse the configuration of hydrogen networks [107]. Whilst not tested on WtH (in literature) this model has the potential to show the possible outcomes both positive and those requiring improvements.

6.2. Practical implications of this study

The results of this study highlight the main issues for the industrial WtH application in real-world scenarios. Along with other studies (Table 1) which provide valuable insight into WtH related technologies, specific investigation into WtH chain and concept is needed. Waste as a feedstock causes technical issues requiring investment, resources and treatment to improve the production yields. Waste pre-treatment adds cost however it improves the purity of the hydrogen increasing the desirability of the product for use in the transport sector. It is essential to provide a hydrogen product to the standards required by the consumer. These challenges will impact future work until costs are in line with other sources of hydrogen and other fuels.

The positive environmental impact, carbon cutting potential, and low GWP provide some of the main promotional values. As a concept that tackles two important issues of reducing waste to landfill or incineration and providing an alternative to fossil fuels. WtH, therefore, needs to be in a position of technical readiness backed by policy targets and support by organisations. Examples of support for clean hydrogen include the IEA report on the future of hydrogen [131], and the European Commission’s European Clean Bus deployment Initiative [132]. Ideally policies, both local and regional, that discuss the next step for zero carbon transport and clean energy would include financial (subsidies) and regulatory support to encourage uptake by companies without undergoing the drawbacks of developing or immature technology.

Following on from this study, research could be extended into; social impact of WtH management systems, hydrogen integration beyond transport, global impact of converting to a hydrogen-based transport

system and innovative cost reduction methods.

7. Conclusions

The process of WtH has a major role to play in the future of providing renewable energy and replacing CO₂ emission intensive fossil fuels, which coincides with the aim of the next generation of clean fuels. The research and analysis so far conducted have shown positive results. Through mixed feedstock gasification process, waste can produce up to hydrogen yields of 33.6 mol/kg and hydrogen concentrations of 82%. While biochemical methods can produce hydrogen up to 418.6 mL/g from fermentation techniques. Energy efficiency techniques such as torrefaction of feedstock can improve the thermal efficiency of gasification up to 4 MJ/kg. This type of enhancement and improvement to hydrogen yields and production rates are related to feedstock processing and advanced energy efficiency processes. Further progress relies on investment, acknowledgement and support for the need to replace fossil fuels with a low carbon alternative, as well as the double benefit of utilising waste and converting it to a valuable resource. The importance of decarbonising the transportation system, finding a zero-emissions fuel replacement for petrol diesel and natural gas has been highlighted. The rate of change and development has been slower than could be argued for the benefits of WtH. Possible explanations include competition from the petroleum industry and the slow-moving speed of cost reductions.

Bottlenecks in technology and management systems hinder the development of the WtH chain. High costs of production and operations, inconsistent feedstock, low efficiencies, inadequate management and logistics, and policy support are some of the bottlenecks discussed. Waste to hydrogen should be part of a combined solution for future energy needs, clean hydrogen production techniques and expected increases in waste linked to population increases and economic development.

Declaration of competing interest

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

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