ACS Sustainable Chemistry & Engineering

pubs.acs.org/journal/ascecg

Research Article

The Environmental Performance of Mixed Plastic Waste Gasification with Carbon Capture and Storage to Produce Hydrogen in the U.K.

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Cite This: https://doi.org/10.1021/acssuschemeng.2c05978

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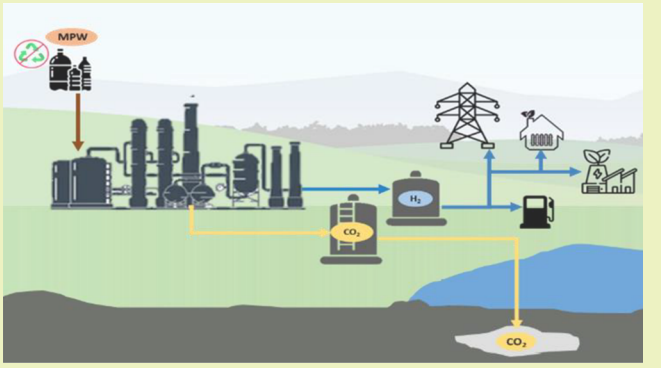
ABSTRACT: Hydrogen of a high purity can be produced from the advanced gasification of nonrecyclable mixed plastic wastes (MPW). Due to the fossil nature of MPW, carbon capture and storage (CCS) capabilities need to be employed for the process to be considered a low-carbon hydrogen production route. This study analyzes the environmental performance of a semicommercial process that (a) provides an end-of-life (EoL) for MPW, (b) produces hydrogen as the main product (for sustainable manufacturing, heating, and transport applications), and (c) captures carbon dioxide emissions which are injected into geological sites for permanent sequestration. The climate change impact result is −371 kg CO_2 per 1 tonne of MPW treated. The process was competitive against a similarly modeled Waste-to-Energy (WtE) plant coupled with CCS—an alternative future end-of-life scenario. WtE with CCS produced a corresponding impact of 17 kg CO_2 per 1 tonne of MPW. The two technologies were also compared alongside a decarbonizing electricity grid mix.



KEYWORDS: *life cycle assessment, waste to energy (WtE), incineration, nonrecyclable plastics, advanced thermochemical treatment*

INTRODUCTION

The tremendous societal benefits and resultant high demand for plastics in the last century have unsurprisingly come at the cost of equally ubiquitous and long-lasting plastic waste accumulation in the ecosphere—a consequence of the same resilient properties that they are revered for.

In 2020, waste arising from plastic packaging in the U.K. totaled ∼2.5 million tonnes, of which ∼47% was mechanically recycled.[1] Therefore, a large volume of nonrecyclable plastic waste, mainly composed of polyethylene (PE) plastic films, polypropylene (PP) food containers, polyvinyl chloride (PVC) pipes, etc., is being directed to alternative end-of-life fates other than mechanical recycling such as incineration (with or without energy recovery) and landfill. Waste-to-energy (WtE) via incineration has been to date the most desirable nonrecyclable plastic waste disposal method, saving landfill space and utilizing the high calorific content to generate heat and electricity, thereby displacing primary energy production and thus virgin fossil fuel. Modern incinerators, however, still suffer from low efficiency and the release of toxic and persistent organic air pollutants such as dioxins, furans, mercury, and polychlorinated biphenyls (from burning of PVC), nitrogen oxides (NOx), and sulfur oxides (SOx), posing a threat to land and marine environments and human health.[2] Typically, PE and PP will result in high polycyclic aromatic hydrocarbon (PAH) levels in the flue gas.[3,4] Another issue is that, with the U.K.'s shortfall of domestic plastic incinerators, the majority of nonrecyclable plastic waste destined for incineration is exported. For example, it is estimated that ∼60% plastic packaging is exported for incineration, primarily to developing countries with inadequate environmental regulations.[5]

This highlights the opportunity for advanced chemical conversion technologies to divert nonrecyclable plastic waste away from incineration and have them treated locally for the production of more high-value products that are not limited to energy generation. Advanced thermochemical treatments of plastic waste, such as gasification, are capable of decomposing nonhomogenous waste into a clean syngas stream providing product flexibility for subsequent upgrading into high-value fuels or chemicals.[6−8] A potential energy dense product from this process is hydrogen.[9−11] With no emissions associated with its point of use and low-carbon production routes available, hydrogen can be a clean energy vector, and thus the development of a low-carbon hydrogen sector is a key element

Received: October 5, 2022
Revised: January 24, 2023

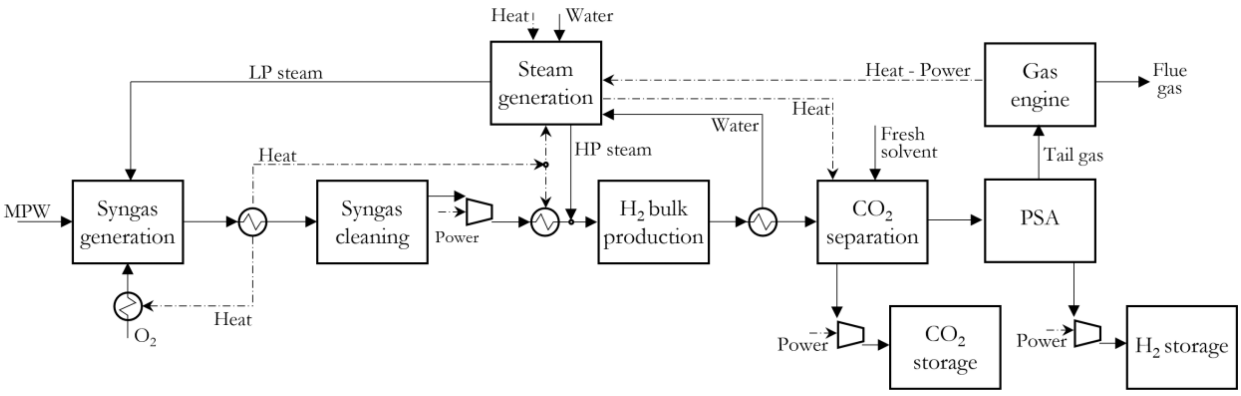


Figure 1. Schematic detailing the H_2 w/CCS process.

of the U.K.'s net zero plan, particularly for "hard-to-decarbonize" sectors like domestic heating, transportation, and heavy industries.[12,13] The use of hydrogen for district heating in the U.K., replacing fossil-based natural gas, is already being facilitated through the "Iron Mains Replacement Programme", and by 2032 the majority of the gas network will be replaced with polyethylene pipes, which are compatible with hydrogen.[14] A demonstration project to blend up to 20% (vol.) hydrogen in the gas network is underway.[15] The H100 project in Fife is planning to supply 300 homes with 100% low-carbon hydrogen through a purpose-built gas network from 2022.[16] Attention, however, must be brought to recent research pointing to the indirect greenhouse effect resulting from fugitive hydrogen emissions to the atmosphere.[17,18] Transport demand will be driven by sectors that cannot be easily electrified such as heavy-duty vehicles or shipping. Industrial users such as refineries, ammonia producers, or glass manufacturers are also seen as early adopters. The Energy Research Partnership estimates current hydrogen demand in the U.K. is 27 TWh/year.[19]

Current commercial scale hydrogen production is dominated by steam methane reforming of fossil fuels (~96%), mainly natural gas.[20] To be considered low-carbon (i.e., Blue-H_2), significant direct CO_2 process emissions will need to be captured and permanently sequestered from the process.[21] Similar carbon performance can also be realized via gasification of different waste feedstocks when coupled with CCS. For feedstocks containing biomass, carbon capture effectively removes biogenic CO_2 from the natural carbon cycle, with the resulting technology considered as a "negative emissions technology" according to the IPCC 2006 guidelines for greenhouse gas accounting.[22] For plastic waste feedstocks, the absence of biogenic carbon means that carbon capture can at best make the technology carbon neutral. Plastics have a higher heating value (20 and 40 MJ/kg) and hydrogen content compared to biomass or municipal solid waste (MPW), resulting in potentially higher hydrogen production.[23]

Gasification as an end-of-life strategy for MPW has been explored previously from a life cycle perspective.[24,25] Life cycle assessments (LCAs) of hydrogen production through this route are less extensively explored, with Midilli et al.[10] calculating preliminary greenhouse gas (GHG) emissions for reviewed lab-scale studies. For biomass and MSW feedstocks, more in depth LCA studies are available with some studies also considering CCS.[26−30] However, a clear understanding of the potential carbon footprint and environmental impact of a plastic-to-hydrogen plant with CCS is not available. This study aims to provide a first comprehensive environmental performance analysis of gasification of MPW to H_2 coupled with CCS. Since the integration of conventional WtE technologies with CCS is also touted as a future technology to improve the sustainability of current WtE plants, the study also provides a comparison against this future waste disposal route.[31,32]

The main goals of this work are

- Highlight the potential environmental benefits of a MPW-to-H_2 gasification with the CCS plant against the backdrop of the current disposal route via incineration
- From a future waste disposal perspective, compare MPW-to-H_2 with CCS to MPW-to-Energy (WtE; incineration) with CCS on an environmental impact basis

TECHNOLOGICAL ASPECTS

The following technologies were modeled using ASPEN Plus for a reference plant treating >35,000 tonnes of MPW per annum producing approximately 7,500 t/a or 470 GWh of hydrogen. This scale is compatible with the output from a reasonably sized Material Recycling Facility (MRF), accounting for residual plastic waste arising from domestic, commercial, and industrial waste. For example, Grundon MRF near Heathrow airport has a capacity of 400,000 t/a of dry mixed recycling waste, ~5−10% of which is nonrecyclable MPW. Process model schematics for the two processes described in this section are provided in Figure 1 and Figure 3.

MPW Gasification for Hydrogen Production with CCS (H_2 w/CCS). The waste generated by Heathrow airport was considered in this work as a case-study reflecting a U.K. scenario. Details for composition and associated analyses are shown in Table 1. Waste collected across all five terminals at Heathrow Airport is sent to a Material Recovery Facility (MRF) where a majority of metals (ferrous and nonferrous), glass, paper and cardboard, and recyclable plastics are normally recovered. The remaining nonrecyclable plastic waste residue from the MRF is dirtied by some residual organic components that adhere to the plastic (such as food remains and paper labels).[33] This fraction of contamination is dependent on the MRF and the inclusion of further cleaning or treatment stages of the residual waste, which may not be included due to added cost and energy for a waste stream that is directed to a landfill or incinerated. The biogenic fraction of this nonseparated, nonrecyclable waste can vary, for example, a 12% and a 23% organic fraction in different samples of rejected plastic waste.[34,35]

https://doi.org/10.1021/acssuschemeng.2c05978
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Table 1. Dry Mixed Recycling (DMR) Composition at the Material Recovery Facility (MRF)

Material Category	Dry Mixed Recycling [DMR] (wt %)
Paper and Cardboard (cardboard, newspapers and magazines, mixed paper, paper cups, beverage cartons)	73.2
Plastic (bottles (polyethylene terephthalate/high-density polyethylene), rigid plastics, flexible plastics)	8.4
Metal (steel cans and aerosols (ferrous), aluminum cans (nonferrous))	2.1
Glass	0.8
Organic (food waste)	4.3
Textiles	0.2
Waste from Electrical and Electronic Equipment (WEEE)	0.0
Printer Cartridges	0.0
Decanted Liquid	3.5
Residual Waste	7.3

Table 2. Mixed Plastic Waste (MPW) Composition Analysis

Component	Mixed Plastic Waste [MPW] (wt %)
LDPE	25
HDPE	25
PP	39
PVC	1
Residual Biomass	10
Ultimate Analysis [wt %]	MPW
C	81.92
H	11.89
N	0.36
S	0.06
O	5.21
Cl	0.57
Proximate Analysis [wt %]	MPW
Moisture	0.95
Fixed Carbon	1.71
Volatiles	95.44
Ash	1.90
Energy Content [MJ/kg]	MPW
Calorific Value (HHV)	41.11

The resulting MPW stream obtained, detailed in Table 2, is transported via trucks to the advanced thermochemical treatment plant where it is incinerated. In this work, the same feedstock will be treated to produce hydrogen, while capturing the CO_2 on site for offshore storage. A distance of 50 km is assumed from MRF to the plant, approximated from the nearest existing incinerators to Heathrow Airport.[36]

The design of the Waste-to-H_2 plant is based on that of more advanced demonstration plants which are at the technology readiness level (TRL) close to commercialization. A process flow schematic is provided in Figure 1. Most of these plants utilize a steam-oxygen blown fluidized bed gasifier operated at 700−800 °C to successfully gasify polymeric chains down to a syngas stream. The raw syngas, containing mostly hydrogen, carbon monoxide, carbon dioxide, and a variety of hydrocarbons including problematic tars, is further treated in a tar-reformer, in this case powered by thermal plasma, at 1200 °C to separate solid particulates and ashes from the stream, while reforming the tars into additional useful syngas.[37] Gasification parameters including temperature of gasifier, equivalence ratio, and syngas composition are in line with those in the literature for MPW gasification.[10] The exiting hot syngas is cooled (down to 200 °C), and the heat is recovered and recirculated to the energy intensive CCS stages. The cooled syngas is cleaned and conditioned using dry filters, acid scrubbers, and alkali scrubber to remove contaminants such as heavy metals, sulfur, and chlorine.[38] The possibility of removing these contaminants from a gas phase rather than liquid makes gasification a more preferable option for chemical recycling of contaminated waste, if compared, for example, to pyrolysis.[39] Following cleaning stages, the gas is fed into a series of water gas shift (WGS) reactors to increase the concentration of H_2 and CO_2. The above-mentioned stages are well established in the chemical industry and have been modeled previously with detailed technological aspects provided in the work by Amaya-Santos et al.[27] The H_2 and CO_2-rich gas is next fed into a conventional precombustion carbon capture unit, comprising of an absorber and a stripper, wherein CO_2 is selectively absorbed in the former at 1.4 bar. An aqueous solution of 30% wt. monoethanolamine (MEA) is used as the solvent. CCS using MEA is a high TRL technology with proven integration for various applications and thus comes with easily transferable knowledge to the process modeled here.[40,41] The stripper subsequently strips the CO_2 out of the liquid solvent using steam, allowing the lean amine to be recycled back to the absorber. Small amounts of circulating amines are sent to the dry filtration unit before discharge; fresh MEA is periodically added to replenish lost solvent. The system employs a 90% carbon capture removal rate and yields a CO_2 stream with a high purity of 99.8%, well above the requirements for grid injection.[42] From the absorber unit, the remaining product gas, stripped of CO_2, is fed to a PSA whereby H_2 of a high purity (99.9%) and low CO concentration (>300 ppm) is obtained according ISO 14687 specifications for H_2 use in fuel cells.[43,44] The hydrogen is pressurized at 200 bar and stored. Remaining tail gas is used to generate electricity via a Jenbacher gas engine. Details pertaining to PSA and gas engine can be found in the work of Amaya-Santos et al.[27]

The dehydrated and compressed CO_2 at 60 bar is transported to the nearest carbon capture and utilization (CCUS), Humberside, from a plant in the Greater London region (Figure 2). The CO_2 transport entails initial lorry transport to the nearest port, assumed to be around 50 km. The CO_2 is then transported via shipping tankers over 500 km from the Port of London to Humberside.[45] From the CCUS cluster, the CO_2 is transported 200 km via pipelines into the North Sea where it is then injected into a deep saline aquifer for permanent storage.[45] The CO_2 is repressurized from 60 to 120 bar to account for pressure drops during pipeline transportation.

Incineration of MPW with CCS (WtE w/CCS). Recently, waste incineration integrated with carbon capture and storage (CCS) technologies has been garnering attention to conciliate climate change concerns.[31,32] However, this integration may suffer from a large energy penalty associated with CCS and lower power plant efficiencies.[46,47] Currently, WtE w/CCS is being deployed at scale in Europe for municipal solid waste (MSW) feedstock (many using amine CCS technologies) and is thus an important comparative scenario for MPW waste disposal. The Twence WtE plant for MSW in The Netherlands will have a CO_2 capture capacity of 100,000 t/yr.[48] At the Klemetsrud WtE facility in Norway, following a successful pilot

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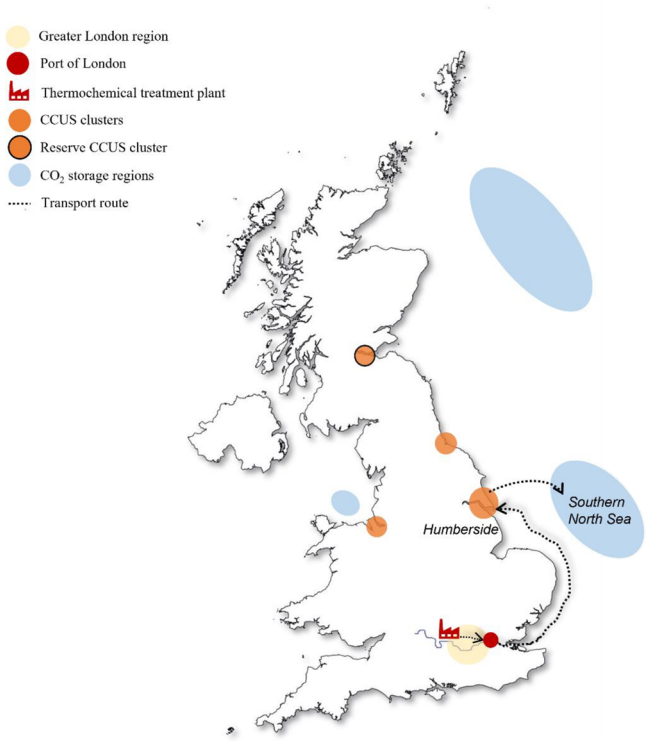

Figure 2. Map of the United Kingdom detailing transport distance for captured CO_2 by freight from Port of London to the Humberside CCUS cluster, transport via pipeline to nearest CO_2 storage region in Southern North Sea and other CCUS clusters and storage regions (adapted from Murugan et al.[42]).

project to capture 90% of direct CO_2 process emissions, development of a full-scale MEA based carbon capture of 400,000 tCO_2/yr is underway.[41] Amager Bakke WtE in Copenhagen, Denmark, has also established a pilot plant.[49]

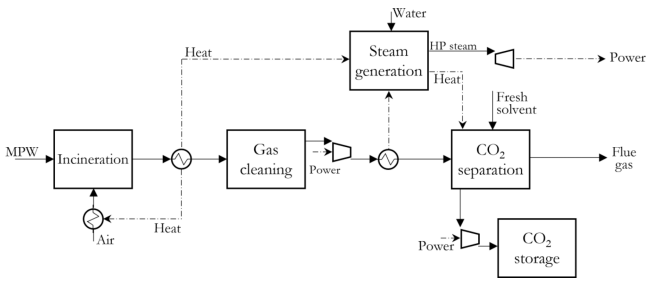

Figure 3. Schematic detailing the WtE w/CCS process.

The process flow schematic is depicted in Figure 3. The modeled WtE plant effectively combusts MPW in an incinerator at >1500 °C in air. The resulting hot flue gas is composed, on average, of 8−10% H_2O, 7−9% CO_2, 75% N_2, and 7−9% O_2, which lies within the typical range for existing WtE plants with some minor deviations due to the difference between MSW and MPW such as moisture and ash content. The resulting hot flue gas generated postcombustion is sent to a heat recovery section which employs a grate-boiler, where high pressure steam is generated. This steam is then used for electricity and heat generation with 8 t/h and 49 t/h of steam generated, respectively, for the scale corresponding to treatment of 1 tonne of MPW (Functional Unit—See Life Cycle Assessment (LCA) Methodology). The flue gas is then treated to remove contaminants using activated carbon, lime, and urea (selective noncatalytic reduction).[50] Additional gas cleaning components were not modeled for the retro-fitted CCS case, although additional cleaning may be required so as to preserve MEA stability and longevity.[51] Instead, a MEA solvent degradation rate was considered to take this into account.[52] In a conventional WtE plant this flue gas is then released to the air. For the retro-fitted WtE w/CCS, the flue gas is instead fed to a carbon capture unit using MEA as absorbent at 1.4 bar where 90% carbon capture rate is achieved. The core carbon capture technology modeled is the same as H_2 w/CCS above; however, it is a postcombustion CO_2 capture rather than the precombustion capture technology for H_2 w/CCS. The advantage of postcombustion is that it can be retrofitted to pre-existing WtE plants and can benefit from long dated experience of CCS from fossil-based power plants.[53] The lean flue gas produced, depleted in CO_2, is released to the environment while the CO_2 captured is compressed, transported, and stored as in H_2 w/CCS.

LIFE CYCLE ASSESSMENT (LCA) METHODOLOGY

The study complies with the ISO 14040 and ISO 14044 guidelines and is modeled on GaBi 10.0.0.71 using Thinkstep and ecoinvent v3.6 databases.[54,55] Primary data is obtained primarily from ASPEN plus modeling of the technologies and is corroborated by plant data from a U.K.-based waste gasification company and waste incineration company.

Goal and Scope. The goal of this work is to assess the environmental performance of managing MPW disposal via gasification coupled with MEA-based carbon capture for the production of high-purity (99.9%) hydrogen. A thorough attributional LCA is conducted for the proposed MPW-to-H_2 with CCS plant, including a hotspot analysis and a consideration of the counterfactual case, namely, diversion of plastic waste to a conventional incinerator. Additionally, a comparative analysis between MPW-to-H_2 with CCS and MPW-to-Energy with CCS is shown, representative of future waste disposal technologies. The functional unit corresponds to the treatment of 1 t of MPW.

The study considers a complex system that (a) utilizes a waste feedstock, (b) produces hydrogen as the main product (for heating and transport applications), (c) captures and permanently sequesters carbon dioxide, and (d) generates electricity. Following the aforementioned ISO standards, a system expansion approach is applied to account for this multifunctionality and thus the avoided environmental impacts of producing electricity and hydrogen are credited to the system.[54,55] Electricity replaces the current U.K. grid mix. As hydrogen is currently not employed at any considerable scale for manufacturing, heating, or transport in the U.K., the study assumes that it replaces natural gas for district heating and is used as the reference scenario throughout this work. This would be similar for manufacturing applications, where natural gas is typically used for heating purposes in gas-fired boilers. It should be noted that hydrogen produced by this process is of transport-grade quality, yet analysis of this use case is omitted due to challenges in conducting an LCA on hydrogen-fueled zero emission vehicles due to technological nascency. To explore the inherent uncertainty related to the choice of the avoided process used for crediting, the study also considers the commercial process for hydrogen production, namely, steam methane reforming (see Introduction). A 10% biogenic carbon

https://doi.org/10.1021/acssuschemeng.2c05978
ACS Sustainable Chem. Eng. XXXX, XXX, XXX−XXX

content is also assumed for the MPW feedstock (which maintain a level of contamination with food and paper labeling), with a ±5% variation. Additionally, a zero-burden approach is applied where any processes associated with plastic prior to it becoming waste is not accounted for. Since the LCA results are also sensitive to impacts and credits for electricity generation, future energy system scenarios for the U.K. in 2030 and 2050 are analyzed for H_2 w/CCS and WtE w/CCS.

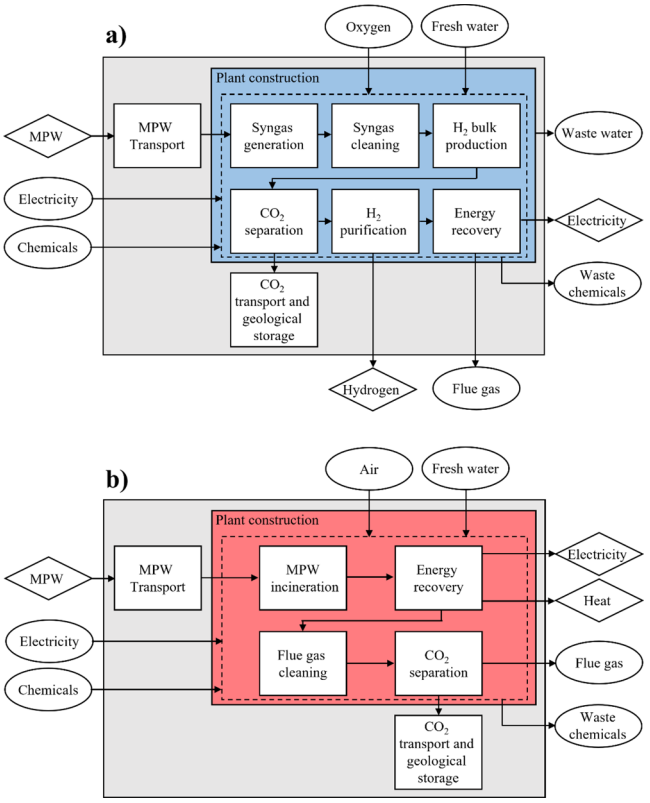

Figure 4. System boundary for the (a) H_2 w/CCS and (b) WtE w/CCS plants.

The system boundaries analyzed are shown in Figure 4. The boundary for MPW-to-H_2 with CCS begins with the transport of the unrecyclable plastics from the MRF to the thermochemical treatment plant. This is assumed to be a distance of 50 km transported via lorry. The processing stages include syngas generation, syngas cleaning and conditioning, carbon capture, hydrogen purification, and compression. The CO_2 captured is transported via lorry and sea tankers and finally through pipelines prior to being injected in a saline aquifer. A similar system boundary is also shown for WtE w/CCS in Figure 4. Life cycle impacts were assessed across the categories that represent the highest environmental priorities according to normalization using the EF 3.0 global reference normalization and weighting factors.[56] The results for all impact categories are reported in the Supporting Information. Hauschild et al. provides a detailed description of these impact categories.[57]

Inventory Data. Data for the Foreground system is obtained from detailed mass-and-energy balances generated via Aspen Plus simulation and are reported in the Supporting Information. Table 3 reports the key inventory data for H_2 w/CCS and WtE w/CCS with respect to the functional unit, 1 tonne of MPW. The environmental burdens of H_2 production include the direct burdens allocated to all the operational units and elementary flows considered in the system boundaries; the indirect burdens allocated to the external supply of material and energy processes; and the avoided burdens allocated to the production of hydrogen, production of electricity from tail-gases, and permanent sequestration of biogenic carbon contaminants. Activities in the background system, which include the provision of materials and energy and the treatment of end-of-life wastes, are modeled using the ecoinvent database, cutoff system model, version 3.8.[58,59] These include the chemicals production and supply required as fluidizing agents (e.g., oxygen); gas cleaning chemicals and CCS solvent (MEA); net thermal energy and electricity requirements/generated; the end of life of ash and MEA discharge via inertization and landfilling; and the treatment of wastewater effluents. Ecoinvent data sets were also used for CO_2 transportation via lorry and sea tankers. CO_2 transportation via pipeline and injection into deep saline aquifers were modeled based on inventory data from Antonini et al.[60] The construction of the plant was also included, using a chemical organics factory from ecoinvent as a proxy and assuming 30 years operation at 80% capacity. Where applicable, data was chosen specific to the U.K. or the Europe region. The gas cleaning stages are based on industry standards, and CO_2 is the main constituent of tail gas emissions. Different scenarios for crediting the avoided impacts from hydrogen production are considered (data retrieved from ecoinvent); the replacement of natural gas in district heating (a conservative approach, as higher grade H_2 is produced here) and the replacement of producing high-purity hydrogen via steam methane reforming of natural gas. The fugitive emissions of hydrogen, a consequence of its application in district heating, is not included, although they impart an indirect

Table 3. Key Inventory Data for H_2 w/CCS and WtE w/CCS[a]

		H_2 w/CCS (90% CCR)	WtE w/CCS (90% CCR)
Inputs			
MPW feed	kg	1000	1000
	MJ	41040	41040
Oxygen (from ASU)	kg	886.5	–
Air	kg	–	19971
MEA solvent makeup	kg	0.7	6.0
Net thermal energy required	MJ	8568	828
Net electricity required	MJ	3780	–
Outputs			
Hydrogen	kg	215.6	–
	MJ	30600	–
Hydrogen purity	%	99.9	–
CO_2 captured	kg	2635	2670
CO_2 purity	%	99.8	99.7
Net electricity exported	MJ	0	2952
Waste-to-X efficiency (energy basis)	%	74	13
CHP efficiency (w/o CCS)	%	70	40
CHP efficiency (w/CCS)	%	57	7
CCS energy efficiency	MJ/kg CO_2 captured	3.68	4.25

[a]ASU: air separation unit. X: valuable products. Further details can be found in the Supporting Information.

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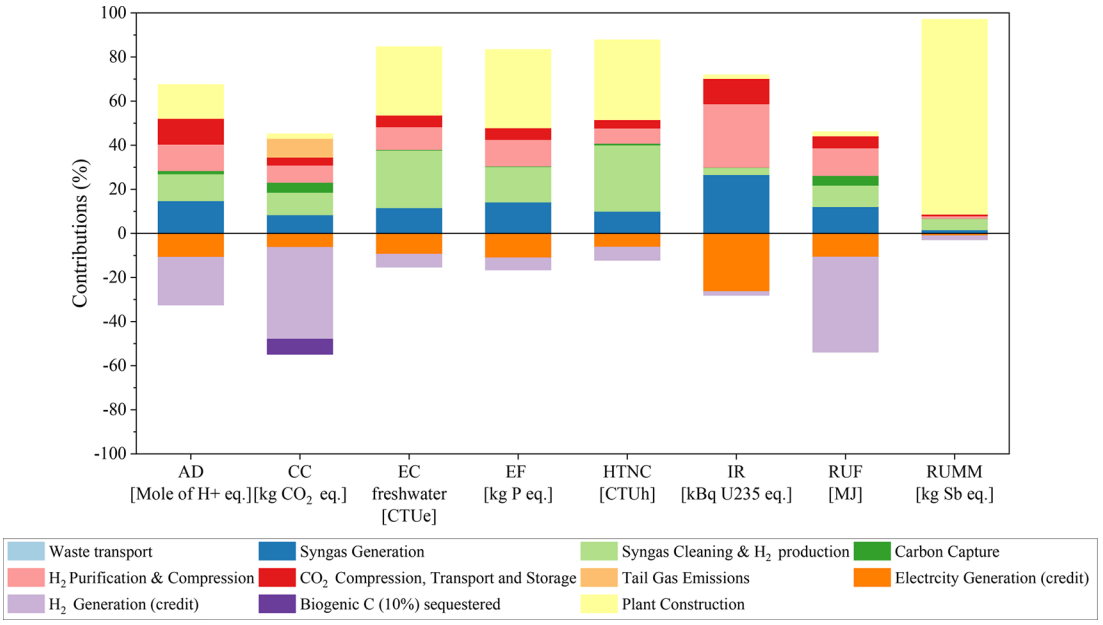

Figure 5. Hotspot analysis results for the H_2 w/CCS plant for Acidification (AD), Climate Change (CC), Ecotoxicity (EC), Eutrophication—freshwater (EF), Human toxicity—noncancer (HTNC), Ionizing radiation—human health (IR), Resource use—fossil (RUF), and Resource use—mineral and metals (RUMM).

impact on climate change; this is a limitation of this study.[17,18] However, for the data set for natural gas for district heating, these emissions are considered. For the scenario analysis, the electricity grid mix for the U.K. in 2030 and 2050 is modeled using the GaBi database.[61,62]

RESULTS AND DISCUSSION

Hotspot Analysis of MPW Gasification for Hydrogen Production with CCS (H_2 w/CCS). Figure 5 shows a hotspot analysis of different stages of a H_2 w/CCS plant for the 8 categories with the highest magnitude of impact after normalization, namely, Acidification (AD), Climate Change (CC), Ecotoxicity (EC), Eutrophication—freshwater (EF), Human toxicity—noncancer (HTNC), Ionizing radiation—human health (IR), Resource use—fossil (RUF), and Resource use—mineral and metals (RUMM).

Waste Transport. Across all categories, waste transport carries negligible (<1%) impact contributions primarily due to a short transport distance and lower mass carrying load (compared to, e.g., CO_2 transport). ***Syngas generation.*** The gasifier and tar reformer have noticeable contributions to all categories analyzed; AD (14%), CC (8%), EC (11%), EF (14%), HTNC (10%), IR (24%), and RUF (12%). The impacts are primarily driven by the energy intensive tar reformer, which consumes ~615 kWh/tonne of MPW (reported electricity requirements range from 400 to 845 kWh/tonne MSW) with >65% of the impacts in the syngas generation section (in categories AD, EC, EF, IR, and RUF) attributed to the tar reformer.[63] Notably, the parasitic power of the tar reforming technology (plasma reformer) is dictated by the amount of ash to vitrify and thus is lower for MPW compared to MSW. For the categories CC and HTNC, the supply of oxygen for the gasifier also contributes significantly as a result of the air separation process.[64] The consumption of oxygen for plastics is higher compared to a biomass or MSW feedstock because plastics have lower content of O_2. ***Syngas cleaningand H_2bulk production.*** The production of chemicals in the gas cleaning stages are substantial contributors to AD (13%), EF (18%), HTNC and (31%). Hydrogen production in the water gas shift (WGS) units requires a thermal and electrical energy input and thus impacts the origin of the supply of these energy systems contributing to CC (9%), IR (8%), and RUF (9%). EC (27%) is contributed nearly equally by chemical supply and electricity. Contributions to cleaning stages have been explored further in Amaya-Santos et al.[27] The syngas cleaning stage also includes the treated landfill disposal of air pollution control (APC) residues and bottom ash, with a negligible impact contribution. ***Carbon capture.*** The stripper solvent regeneration unit constitutes the most thermal energy intensive unit of the entire plant requiring 2.7 MWh/tonne MPW even with some thermal energy recovered in the syngas generation stage used to offset this demand. The associated impacts, CC (7%) and RUF (7%), are significantly reduced by using internally generated heat. Although total volumes of MEA solvent are large, fresh makeup solvent is also considered and does not impart a large impact. ***H_2 purification and compression.*** The electricity required to pressurize the H_2-rich stream to 20 bar for PSA operation is the main contributor to most categories: AD (12%), CC (8%), EC (10%), HTNC (7%), IR (27%), and RUF (12%). ***Tail gas emissions.*** CC (8%) impacts originate from 10% of carbon released to the air after capture (with a small part of these emissions with biogenic origin). ***CO_2 compression, transport, and storage.*** Compared to other stages of the process, impacts associated with all categories are low for CO_2 transport and storage with a notable contribution only to AD (12%), 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 CO_2 eq/tkm. The U.K. is well poised to exploit infrastructure and expertise of its expansive gas network and transport waterways.[45] Electricity for compression from 1.5 to 120 bar contributes to IR (11%).

https://doi.org/10.1021/acssuschemeng.2c05978
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Plant construction. Although climate change impact is negligible compared to other stages of the process, construction does contribute to AD (15%), EC (31%), EF (35%), and RUMM (88%) due to large steel and thermal energy requirements. ***Credits.*** Hydrogen production credits for replacement of natural gas for district heating results in savings for AD (−21%), CC (−41%), and RUF (−43%). Similarly, savings across all categories (barring RUMM) ranging from −6% to −25% are associated with electricity generated from tail gas via a gas engine due to its high H_2 content (although electricity is generated, the process still requires net electricity input). Since MPW feedstock is contaminated by 10% biomass, savings in CC (−7%) are shown from sequestration of biogenic C.

Different configurations of this process may impart varying impact contributions. Based on this hotspot analysis, alternative technologies can be explored. The traditional amine solvent-based carbon capture technology could be replaced by other mature technologies such as Selexol, Rectisol, or Benfield.[65] Some lower TRL technologies also show promising energy savings.[66] The H_2 produced is of a high purity for fuel cell use, and thus changing specifications of H_2 for other applications would reduce impacts associated with the PSA and the tar reformer. Other H_2 separation technologies such as membrane separation can be explored.[67]

Climate Change Impact of the H_2 w/CCS Plant. The climate change impact of the H_2 w/CCS plant is presented per FU alongside uncertainties related to feedstock composition, hydrogen crediting approach, and energy efficiency configuration (Figure 6).

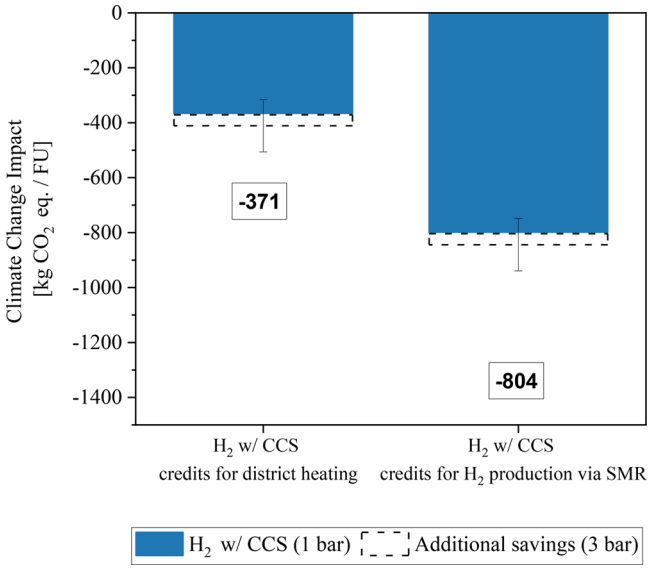

Figure 6. H_2 w/CCS results showing uncertainty from choice of crediting approach, increased energy efficiencies and carbon savings with a different technology configuration, and error bars associated with changes in biomass contamination of feedstock.

The climate impacts for two different crediting approaches to hydrogen, namely, replacement of hydrogen from conventional production via steam methane reforming of natural gas and replacement of natural gas in district heating, highlight the sensitivity of results to hydrogen credits and the large uncertainty that arises from this modeling choice.[68] Both crediting options are reasonable and justifiable. MPW-to-H_2 is proposed as an alternative production pathway to the conventional hydrogen production route of steam methane reforming (SMR) of natural gas (where H_2 is of comparable purity). The total impact considering avoided burdens of either producing hydrogen via conventional means or using hydrogen for district heating are −804 kg CO_2 eq/FU and −371 kg CO_2 eq/FU (or −48% and −42% contributions), respectively. Interestingly, H_2 in the atmosphere may have some indirect warming effects on climate, and a growing body of research is investigating the impacts of fugitive H_2 emissions.[17,18] Future research is directed toward addressing the comparative impacts between methane and H_2 fugitive emissions and avenues to reduce H_2 emissions in future applications.

A baseline scenario of 10% contamination with biomass was modeled based on waste composition data. With the application of CCS, impacts become sensitive to changes in feedstock composition and a ±5% contamination is expected, causing an impact of ±95 kg CO_2 eq/FU. An interplay exists between biomass and plastic feed compositions. Larger calorific value observed for plastic-rich feed leads to greater feedstock/syngas and feedstock/H_2 efficiencies. A biomass feedstock, however, gains substantial environmental advantages due to its biogenic C content which, when coupled with CCS, generates carbon savings and offsets the marginal benefits from increases in H_2 production and lower feedstock mass throughput (thus lower waste transport contributions) for plastic waste.

The more energy efficient scenario of H_2 w/CCS where syngas is pressurized to 3 bar prior to WGS and CCS stages is also presented to show uncertainty in technology configurations. The process introduced in the technical description above was considered as the baseline case for this research. A more energy efficient plant was also modeled, whereby the clean syngas and steam are pressurized separately to 3 bar first. The absorber unit operates at 3 bar. The energy benefits of this are seen downstream at the PSA, whereby a large volume flow is then compressed to 20 bar to utilize the PSA. Savings equivalent to −40 kg CO_2 eq/1 t of MPW are achieved with this configuration (Figure 6).

Counterfactual Analysis for the H_2 w/CCS Plant. The H_2 w/CCS is also compared with different configurations of a conventional WtE plant in Figure 7. The total impact considering avoided burdens of producing hydrogen via SMR for district heating is −371 kg CO_2 eq/FU. The total positive impacts, without the inclusion of hydrogen and electricity credits (thus only regarding internal heat recovery for CCS section), is 1453 kg CO_2 eq/FU. The climate change impacts for two different counterfactual cases are provided. For an incineration plant modeled on ASPEN, considering only internal heat recovery and no electricity exports, 2655 kg CO_2 eq/FU is emitted. This is in line with the ecoinvent data set for a similar feedstock (2682 kg CO_2 eq/FU). The emissions for these cases are dominated by CO_2 emissions to the air. In Europe, a WtE plant with electricity and heat exports is more common and was modeled with an impact of 1617 kg CO_2 eq/FU (notably, a figure similar to that of H_2 w/CCS when credits are not considered). These results highlight the benefits of CCS applied to gasification for waste treatment and the value in producing hydrogen. Therefore, inclusion of the avoided burdens of the counterfactual case would lead to an even greater negative impact than the −371 kg CO_2 eq/FU of the H_2 with CCS system. Its evident that consideration of the counterfactual case can have a significant influence on results.

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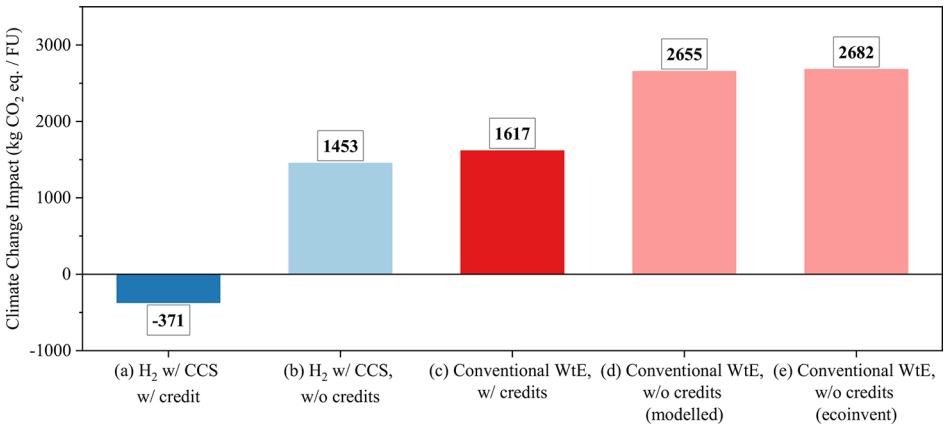

Figure 7. Climate change impact results for H_2 w/CCS against conventional WtE as a counterfactual. The following configurations are presented: (a) H_2 w/CCS, including H_2 and electricity credits, (b) H_2 w/CCS, without H_2 and electricity credits, (c) conventional WtE, including heat and electricity export, (d) conventional WtE, including internal energy recovery, no exports (modeled), and (e) conventional WtE, including internal energy recovery, no exports (ecoinvent).

Comparison with Incineration with CCS. An environmental comparison between a Hydrogen with CCS and WtE (incineration) with CCS plant is conducted as potential future scenarios for disposal of MPW.

For Waste-to-Energy as a waste disposal method to stay relevant in the coming decades, its environmental performance will need to be improved via a postcombustion capture system. Several European projects are underway to retrofit WtE plants with CCS capabilities. In WtE plants, flue gas cleaning technologies to remove particulates, nitrous oxides, and dioxins have improved substantially; however, a large percentage of carbon is completely combusted to CO_2 which is subsequently released to the environment. For a plastic waste feedstock, the WtE plant modeled in this study yielded 2.9 tonnes of CO_2 (fossil and biogenic) for every tonne of MPW combusted. The WtE w/CCS plant is modeled to redirect some high-pressure steam used for electricity generation to provide the heat required for the solvent regeneration at the reboiler. This allows self-sustained operation of the WtE and CCS plant albeit at the cost of electricity generated and supplied to the grid. With the current political climate and unpredictable costs surrounding natural gas supply, this was considered the preferred operational scenario.

Climate change impact results for the two technologies are presented in Figure 8. For both technologies, CCS implementation comes at a significant energy penalty for operation. In comparison to precombustion in H_2 w/CCS, the CO_2 in the flue gas of WtE w/CCS is highly diluted with N_2 resulting in low CO_2 partial pressure (11.9 kPa compared to 44.2 kPa for H_2 w/CCS), higher circulation volumes of MEA, and higher duty to regenerate the solvent at the stripper. Despite the greater thermal energy for solvent regeneration in WtE w/CCS, internally supplied heat from medium pressure steam brings down the climate change impact of the CO_2 section compared to H_2 w/CCS. Similarly, impacts associated with the supply of the solvent, MEA, are higher for WtE w/CCS. The lower energy demand for H_2 w/CCS due to higher partial pressures of CO_2 is balanced by energy requirements upstream for the air separation unit (for O_2 supply), reforming/gasification, and lower heat recovery. These factors along with hydrogen purification and compression stages ultimately lead to a total positive impact around 3 times higher at 1715 kg CO_2 eq/tonne of MPW treated compared to 659 kg CO_2 eq/tonne of MPW treated for WtE w/CCS. Stack emissions of fossil CO_2 are similar between the two technologies.

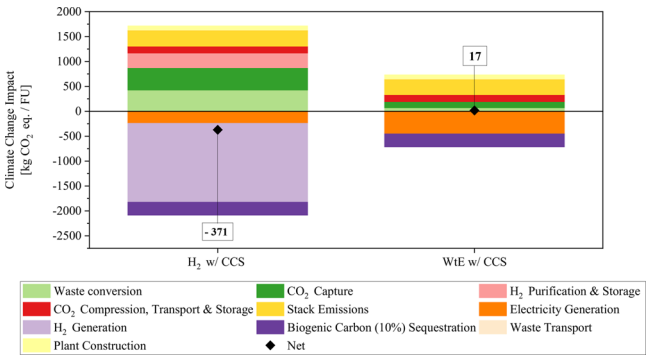

Figure 8. Climate change impact for H_2 w/CCS and WtE w/CCS showing contributions from stages and net total impact. Here, the "Waste conversion" section refers to all stages of the process prior to the carbon capture units.

The impacts associated with the avoided burdens is a crucial differentiator between these technologies as one produces electricity as its main product, while the other produces high-quality hydrogen as its primary product and electricity as a secondary product. The electricity generated in a steam turbine and exported in WtE w/CCS is 1.5 MWh per FU compared to 0.78 MWh per FU generated from flue gas in a gas engine for H_2 w/CSS, and avoided burdens associated with electricity production for the U.K. reflects this with an additional −212 kg CO_2 eq/FU savings for WtE w/CCS. However, considering the avoided burdens for hydrogen from natural gas used for district heating, the total impact for H_2 w/CSS is −371 kg CO_2 eq/FU respectively, compared to 17 kg CO_2 eq/FU for WtE w/CCS. This could be an underestimation of the credits associated with hydrogen due to the high purity of H_2 produced by the process which is not accurately reflected in its replacement of natural gas for district heating. Additionally, avoided burdens are also associated with the permanent sequestration of the biogenic fossil component of waste which corresponded to a 10% biomass contamination of MPW feedstock with minor differences in magnitude between technologies.

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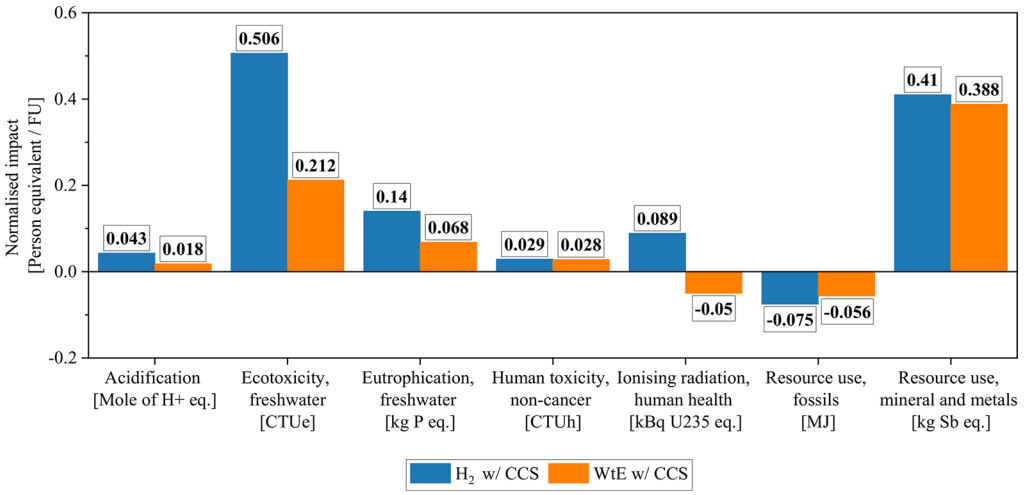

Figure 9. Normalized impact results in person equivalent (PE) per tonne MPW.

In addition, the counterfactual case of conventional WtE (as applied in the Counterfactual Analysis section), can be also considered for the WtE with CCS case. Since the avoided burdens from the counterfactual case will be of the same magnitude for H_2 w/CCS and WtE w/CCS, the relative impacts for comparison between these technologies are the same.

The normalized results, using EF 3.0 method global normalization factors with units "person equivalents", for all other impact categories are shown in Figure 9. Impact categories with the largest magnitudes include Ecotoxicity (freshwater), Human toxicity (noncancer organics), Resource use (minerals and metals), Resource use (fossils), Ionizing radiation (human health), Eutrophication (freshwater), and Acidification. For most categories, differences between technologies can be attributed to the allocation of electricity credits (WtE w/CCS) vs the allocation of hydrogen credits and electricity burdens (H_2 w/CCS). WtE w/CCS fares better in most categories due to net electricity production especially with respect to those dominated by the electricity grid—Ionizing radiation, Eutrophication (freshwater), and Acidification. For ecotoxicity, the oxygen supply and chemicals for cleaning, particularly sodium hydrochlorite, render H_2 w/CCS more environmentally unfavorable than WtE w/CCS, which does not require oxygen and uses different chemicals, particularly urea and lime. A more robust cleaning process is crucial for H_2 w/CCS to yield H_2 of the desired purity. Catalysts during the WGS stages are also susceptible to poisoning. Resource use (fossils) is primarily dependent on avoided burdens from use of natural gas, and the ranking is based on natural gas use intensity associated with the district heating and electricity grid.

Scenario Analysis. Many LCA examples of industrial processes are shown to be strongly affected by changing energy systems, namely, electricity and heat, from both a temporal or regional context.[69,70] Future decarbonization of heating for industrial processes has not been analyzed, as one of the main low-carbon alternatives to thermal energy from fossil fuel combustion is hydrogen (the other being biomethane from sustainable biomass sources). This however does highlight the value in producing hydrogen and the potential for internal use of hydrogen for heat recovery. The evolution of the district heating and other hydrogen production technologies would also serve as interesting scenarios to explore but has been excluded from this work. The climate change scenario results for changes to the electricity supply on H_2 w/CCS (only the conservative district heating case is presented) and WtE w/CCS are shown in Figure 10. The forecasted carbon intensities of the U.K. electricity grid mix in 2030 and 2050 are 0.193 kg CO_2 eq/kWh and 0.178 kg CO_2 eq/kWh, respectively.[62]

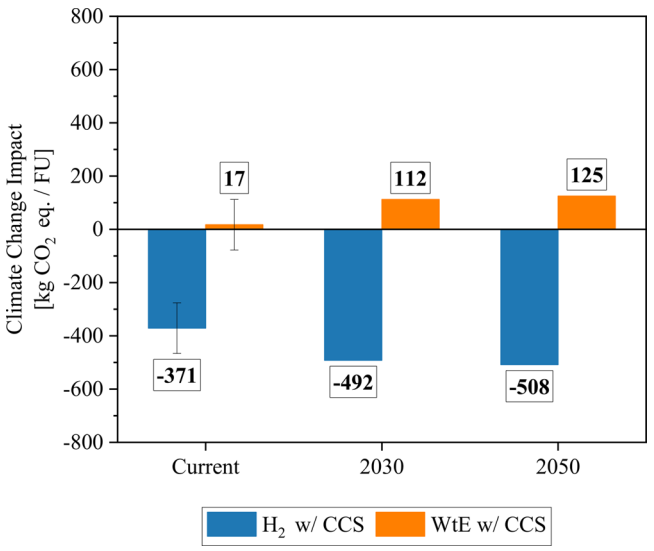

Figure 10. Contribution to climate change for different electricity grid mix scenarios.

H_2 w/CCS shows a reduction by 33% to −492 kg CO_2 eq/FU by 2030 and a 37% reduction to −508 kg CO_2 eq/FU by 2050, relative to the current scenario. This sensitivity to changes in the grid carbon intensity is reflective of the large net electric power of 1.05 MWh/tonne of MPW for the energy intensive stages highlighted in the hotspot analysis previously. Thus, H_2 w/CCS tends toward better environmental efficiencies as electricity becomes less carbon intensive. The opposite trend is observed for WtE w/CCS; the sensitivity to electricity changes is pronounced as electricity requirements and generation are the largest contributors alongside flu gas emissions for WtE w/CCS at 16% and −38%, respectively (Supporting Information). An increase from to 112 kg CO_2

https://doi.org/10.1021/acssuschemeng.2c05978
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eq/FU and 125 kg CO_2 eq/FU is observed for the 2030 and 2050 scenario. According to these results, WtE w/CCS becomes less environmentally beneficial with the implementation of CCS, as credits for electricity begin to carry less weight. This is an important result as it highlights that incineration of plastic even when retrofitted with CCS capabilities may not be a long-term solution for plastic waste disposal, particularly in light of net-zero targets, and thus must be explored further. This is in contrast to gasification or incineration with CCS of a biomass-dominant feedstock that will continue to show climate negative impacts due to the permanent sequestration of a biogenic carbon (BECCS).[27,47] The results for all other impact categories follow a similar trend with those that have greater contributions associated with the electricity grid mix showing more sensitivity.

The conversion efficiencies of electricity production via Jenbacher gas engine (H_2 w/CCS) and superheated steam turbine (WtE w/CCS) are mature technologies, and impacts in the future are unlikely to change drastically on account of improved efficiencies. Also, the operational configuration chosen for WtE w/incineration will impact results because of the interplay between electricity and heat production. The configuration analyzed here pertains to increased heat production at the expense of electricity production to cover all thermal energy requirements of the CCS unit. When the system is adjusted to maximize electricity production, external thermal energy will need to be supplied. However, the substitutional value of electricity generation will still be reduced, leading to positive impacts.

CONCLUSION

It is becoming more pertinent to find a suitable disposal system for nonrecyclable mixed plastic waste (MPW), as incineration (WtE) and landfill are insufficient and outdated methods. In this study, the environmental performance of gasification of MPW as a flexible disposal route is investigated.

The process produces a high efficiency energy vector, hydrogen, as its main product and captures carbon dioxide using an amine-based solvent capture technology (at 90% carbon capture rate) which is subsequently transported and injected for permanent geological storage. High electricity requirements for tar reforming during syngas generation and high thermal energy requirements at the CCS stage are environmentally costly. For applications requiring high-purity H_2, separation and purification of the stream also imparts a high impact from the energy load.

The LCA study adopts a "waste perspective" focusing on the environmental performance associated with the management of waste. In this context, H_2 w/CCS yields a net negative climate change impact of −371 kg CO_2 eq/1 tonne MPW treated when considering credits allocated to the production of hydrogen, assumed as avoided burdens from use of natural gas for district heating. The LCA results are influenced by the choice of the crediting approach for H_2. WtE coupled with CCS is touted as an alternative low-emission disposal method with the possibility of retrofitting existing WtE plants. This configuration yields a net positive climate change impact of 17 kg CO_2 eq/1 t MPW. In addition, the environmental benefits of WtE plants become less marked as credits from electricity generation (which is its high-value export) become less relevant in the changing energy systems landscape. On the other hand, H_2 w/CCS, which is a net electricity consumer, is advantaged by decarbonization of the grid. Similarly, the environmental performance of both technologies is sensitive to biomass contamination of MPW due to permanent sequestration of biogenic carbon.

Advanced gasification with CCS is proposed as a potential EoL management for nonrecyclable mixed plastic waste. In light of net-zero targets, the production of H_2, with technological flexibility in desired purity, is a crucial feature of the process. The treatment of MPW to produce hydrogen (with CCS) could contribute to increasing chemical recycling rates and moving toward a clean hydrogen economy.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssuschemeng.2c05978.

(a) Details on feedstock composition, (b) mass−energy balances for different technologies and configurations, (c) LCA methodology, and (d) life cycle impacts for all categories (PDF)

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

The authors wish to thank Matt Prescott (Heathrow Airport) for providing information related to waste feedstock and ABSL engineers for providing technical data of the gasification plant.

ABBREVIATIONS

AD, Acidification; ASU, Air separation unit; BECCS, Bioenergy with carbon capture and storage; CCS, Carbon capture and storage; CCUS, Carbon capture and utilization and storage; CHP, Combined heat and power; CC, Climate Change; EC, Ecotoxicity; EF, Eutrophication—freshwater; FU, Functional unit; HDPE, High density polyethylene; HHV, Higher heating value; HTNC, Human toxicity—noncancer; IR, Ionizing radiation—human health; LDPE, Low density polyethylene; MRF, Material recovery facility; MJ, Megajoule;

MPW, Mixed plastic wastes; MEA, Monoethanolamine; MSW, Municipal solid waste; NOx, Nitrogen oxides; PE, Person equivalents; PE, Polyethylene; PP, Polypropylene; PAH, Polycyclic aromatic hydrocarbons; PVC, Polyvinyl chloride; PSA, Pressure swing adsorption; RUF, Resource use—fossil; RUMM, Resource use—mineral and metals; Sox, Sulfur oxides; SMR, Steam methane reforming; TRL, Technology readiness level; WtE, Waste-to-Energy; WEEE, Waste from Electrical and Electronic Equipment; WGS, Water gas shift; wt %, Weight percent; vol %, Volume percent

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https://doi.org/10.1021/acssuschemeng.2c05978
ACS Sustainable Chem. Eng. XXXX, XXX, XXX−XXX