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Article

Hydrothermal Upgrading Of Waste Plastics: An Environmental Impact Study

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Abstract: This paper presents the life cycle assessment results of a study of plastic recycling *via* hydrothermal upgrading (HTU), a chemical recycling technology. It was investigated due to its potential to address current gaps in the plastic recycling system, largely due to several plastic packaging materials and formats that cannot be processed by traditional mechanical recycling technologies (primarily due to technoeconomic reasons). As society transitions towards a net-zero-based circular economy, assessments should be conducted with a futuristic outlook, preventing costly mistakes by employing the right technologies in the right areas. HTU can generate up to 76% reduction in climate change impacts when compared with comparable end-of-life treatment technologies whilst conserving material with the system. Additionally, further reductions could be achieved by changes in electricity consumption within the study. This represents a new insight to chemical recycling of polymers by establishing a prospective life cycle assessment study that looks to introduce a step-change in the recycling system. By creating a transparent cut-off model with consistent allocation, it highlights the benefits of introducing this technology as opposed to the current model of disposal through incineration or landfill.

Keywords: hydrothermal upgrading; naphtha; life cycle assessment; chemical recycling; waste plastic

1. Introduction

The phenomenal growth of the plastics industry has resulted in a vast number of plastics produced worldwide every year, contributing to the materialistic affluence in human living (Álvarez-Chávez et al., 2012; Tabone et al., 2010). However, the abundance of plastic goods in the world has also created serious environmental problems. Globally, around 367 million tonnes of plastic waste is produced annually and with predicted plastics demand, the global annual production of waste plastic will almost double by 2030 (Plastics Europe, 2021). The ever-increasing amount of plastic wastes is overwhelming conventional waste management infrastructure – landfill, energy from waste, mechanical recycling - globally. In the UK and the EU, disposal of waste to landfill has been significantly reduced through environmental regulation, driven by market measures and so protect the environment from intractable impacts including hazardous leachate, groundwater pollution and gaseous emissions (U S Environmental Protection, 2010).

Following governmental direction in the UK (HM Government, 2018; WRAP, 2018) EU(European Commission, 2020) and developing countries where plastic is a symptom of economic growth,(Nandini and

Chattopadhyay, 2020) solutions are being sought to address the recycling of currently non-recyclable, single use plastics, but to do so in a way that makes both environmental and economic sense. Recycling post-consumer plastics at scale, following mixed waste collection from the domestic and commercial settings, is a very complex challenge (Lange, 2021; Ragaert et al., 2017). Plastic bottles (PET & HDPE) are extracted and sent to recyclers for closed loop recycling, whereas other plastics, roughly sorted into single polymer bales, are sent for further processing to Material Recovery Facilities (MRFs) or exported for incineration. Polymers which can be sorted to a high degree of purity at MRFs are mechanically recycled using heat and pressure treatments to form flakes or pellets which can be extruded or blow moulded into new uses (Burgess et al., 2021).

Using mechanically recycled plastic in higher-value applications is technically challenging and leads to increased process rejects and a generally lower quality of product, which is why traditional mechanical recycling processes are often referred to as ‘down-cycling’ (Hong and Chen, 2017). It is possible to generate good quality products via mechanical recycling provided there is good control of the quality of the input feedstock. Optimally, mechanical recycling requires clean, homogeneous feedstocks, which will have required sorting, and cleaning to remove grit and food residues, and contamination (paper, card, and other non-target materials). But such processes do not remove embedded colour, separate different polymers or other materials in multi-layer packaging or address deep contamination of the plastic; all of which influence the recyclate end-market and valorisation. Chemical recycling processes are however able to tolerate a wider range of non-homogeneity of post-consumer plastics, relative to traditional re-processors and therefore these offer a practical solution at scale, as part of a circular economy model for plastics (Ragaert et al., 2017). Chemical recycling has been defined as *“any reprocessing technology that directly affects either the formulation of the polymeric waste or the polymer itself and converts them into chemical substances and/or products whether for the original or other purposes, excluding energy recovery”* (Chemical Recycling Europe, 2019).

Hydrothermal upgrading (HTU) combines the process characteristics of pyrolysis (high heat) and solvolysis (dissolution) to heat, melt and then dissolve in steam the mixed plastics feedstocks under supercritical conditions (>373 °C, >220 bar) (Maschmeyer and Humphreys, 2011). In this process environment, polymers are broken down into short-chain hydrocarbons, with the water acting as a hydrogen ion donor to reduce excess cracking and free-radical formation. The steam-hydrocarbon mix is then passed through a column containing a solid-state catalyst and depressurised to flash distil the products out across the range of boiling points. The catalyst is not broken down or consumed in the reaction, nor does it need regeneration. Through the direct transfer of heat to the plastic *via* the supercritical water process environment, there are no obvious limitations to process scale up. Additionally, as it is not a direct-heating combustion process, HTU does not create harmful combustion by-products such as dioxins and does not produce char as a by-product of conversion reactions, and so helps to maximise higher product yields than that of other thermal process technologies. The aim is that through chemical recycling, a step-change in the recycling system will be made to address ‘performance gaps’ left by traditional mechanical recycling technologies, that otherwise directs waste plastic to incineration or landfill. However, there has been some concern that the energy demand associated with chemical recycling, due to the process conditions involved, makes these process more environmentally damaging (Green Alliance, 2020). Moreover, identifying a sustainable pathway to net zero requires operators to consider consumption, process efficiency and renewable energy as options, to ensure that circular systems deliver better environmental outcomes than the conventional systems.

To understand the environmental impact more clearly, life cycle assessment (LCA) must be completed in a consistent manner to be able to compare the benefits and weaknesses of technologies. Previous work has looked at how advanced chemical recycling can contribute to reducing emissions, but without a focus on a specific technology (Agora Energiewende and Wuppertal Institute, 2019; Enkvist and Klevnäs, 2018; Material Economics, 2019). Additionally, existing LCA studies that covered chemical recycling have been reviewed (Davidson et al., 2021) but highlighted there was only a handful of studies in this area. Some other commissioned studies have focused on technological comparisons aiming to analyse the environmental impact of using chemical recycling technologies compared to traditional plastic waste treatment (BASF, 2020; CE Delft, 2019). Furthermore, existing studies in this area have relied on industrial scale production data for one chemical recycling technology (pyrolysis) (BASF, 2020) or others based on demonstration plants that are in use but not yet at commercial scale (CE Delft, 2019). Studies assessing pyrolysis in the UK, Singapore and Europe suggest its climate change impact is significantly lower than that of incineration (Gear et al., 2018; Jeswani et al., 2021; Khoo, 2019; Somoza-Tornos et al., 2020). Whilst academically interesting, it should be noted that chemical recycling is likely best viewed as complementary to existing techniques such as mechanical recycling, rather than a competitor, due to the different quality of feedstocks that it can process.

A pyrolysis study to produce propylene and ethylene monomers in the United States found a significant influence on the climate change impacts due to regional grid mixes (Gracida-Alvarez et al., 2019). Analyses was presented on waste management in countries and European cities and compared with pyrolysis of plastic waste to produce petrochemicals including gasoline, propane and butane (Al-Salem et al., 2014; Alston et al., 2011; Iribarren et al., 2012). The pyrolysis scenarios they examined (futuristic or hypothetical) appear to offer environmental benefits in terms of consumption of fossil fuel resources and GHG emissions compared to some reference scenarios like material recovery, combustion, and landfilling. Other studies have reported pyrolysis of plastic waste as offering more environmental benefits, such as reduction of GHG emissions and consumption of fossil fuels, compared to other plastic waste treatment options (Barry et al., 2019; Benavides et al., 2017; Demetrious and Crossin, 2019; Vienescu et al., 2018).

As can be seen from the literature, there have been several case studies on LCA of plastic waste treatment via pyrolysis but none using HTU. As a result, the aim of this paper is to carry out the first LCA study of recycling plastics into a naphtha-like fraction using relevant industrial data for a scaled-up facility, specifically *via* HTU, to understand its environmental impacts and where potential savings can be made for the future.

2. Materials and Methods

The LCA was conducted following the guidelines contained within ISO 14040/14044 recommendations (ISO, 2006a, 2006b). This is broken down in the following subsections.

2.1. Goal and scope definition

The primary goal of the study is to carry out an assessment of hydrothermal upgrading to understand its impact on the environment. The scope is cradle-to-gate using an attributional cut-off approach, focusing on understanding the potential environmental impact of the activities themselves. The HTU process was modelled on the HydroPRS™ technology and located at a facility which is currently being constructed at a site in Wilton, UK. As such, this is a pre-operational assessment based on conservative estimates for the process, using a standardised approach of estimating the highest potential usage or consumption of the relevant inputs.

For this study, the waste feedstock materials will be assumed to have zero environmental impact (as the burden is assumed to lie with the primary production) and the focus will be on the recycling operations itself. The boundary will start at the journey of the material to the Wilton site and ends with the production of the five product streams generated by the HTU process. Electricity was sourced from a mix of 16% biomass and 84% UK grid mix, as is expected to be supplied to the Wilton site, Teesside UK.

2.2. *Functional unit, data collection and allocation*

The functional unit for this study is the processing of one tonne of waste plastic feedstock. Primary data for the HTU process was supplied by ReNew ELP based on projections from the facility design stage. As this is a process to produce material feedstocks for the chemical industry, a mass-based allocation approach was taken with the co-products. A procedure based on allocation by economic value was considered but not progressed due to the prospective nature of the assessment and the true economic value of each of the products not being fully realised at this stage, as well as the changing economic conditions that were being experienced during the development of the work. GaBi software was used to create the LCA models and generate impact assessments using the ReCiPe 2016 methodology (hierarchical viewpoint).

2.3. Feedstock Treatment

The overall model is effectively a cradle-to-gate process, where the environmental burden is allocated from the start of the journey of the polymer feedstock to the Wilton site. This is assumed to be 160 km by a 34-40 tonne (gross weight) Euro 6 standard truck; the distance was defined based on the likely polymer feedstock supplier. The expected composition of this feedstock was determined based on discussions with ReNew ELP and their suppliers and assigned to an end-of-life pathway to assist with the LCA development. As there are no quality standards within the UK for waste plastic bales, the supplied plastic waste feedstock is expected to contain non-target contaminants which need to be removed within a purpose-built material preparation plant (MPP) before the polymeric material can be processed within the HTU system. The feedstock is shredded, and metal contaminants are removed at this first stage. A dry clean of the shredded feedstock removes further solid contaminants such as stones, glass, wood, grit etc. These contaminants are expected to be typically 23 kg of inert waste (which will be transported 10 km by truck to a landfill), and 8 kg ferrous metal & 2 kg non-ferrous metals (which are sold to other recyclers). The final stage of the cleaning process is a near-infrared sort to ensure only the appropriate polymers enter the process. Rejects here include PVC, paper, dry cleaner dust etc. with a total mass of 72 kg. They are all combustible and are expected to be transported 18 km by truck to an energy-from-waste facility. Emissions from the EfW facility have not been attributed to the HTU process as that activity would have occurred regardless of a chemical recycling intervention, and thus it is not fair to attribute the activity in this instance. The remaining 895 kg of waste plastic (predominantly LDPE & PP) would be processed *via* hydrothermal upgrading.

2.4. *Hydrothermal upgrading*

Hydrothermal upgrading (HTU) is effectively a one-stage process (Figure 1) with several different inputs as listed below. As the catalyst is not consumed within the reaction, it is not considered as an input for the process as the overall environmental impact per tonne of input is neglible.

- Electricity usage: 16 200 MWh/annum
- Consumption of potable water (fire water, safety showers and general hygiene): 900 tonnes per annum
- Demineralised water used in the boiler: 14,000 tonnes per annum
- Process water used to enhance the cooling of the system, particularly during the summer: 8,500 tonnes per annum
- Process gas used onsite for steam generation: 1600 tonnes/annum

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- Wastewater treatment used to remove contaminants from the wastewater: 15,768 tonnes per annum

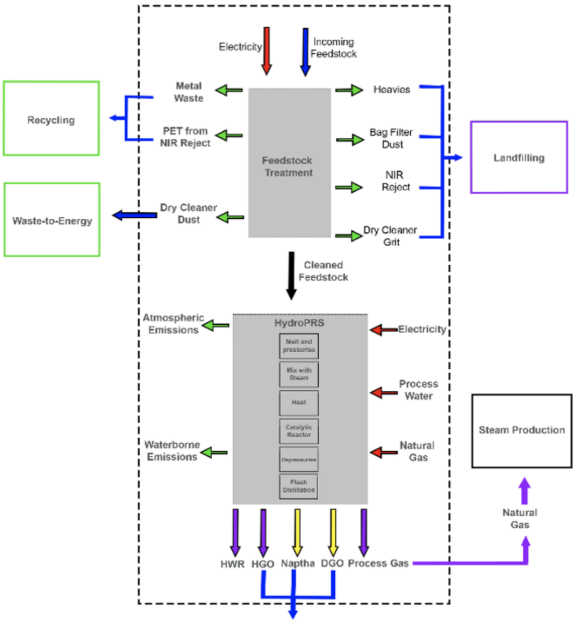

Figure 1 - Schematic of the material preparation and hydrothermal upgrading process

This inventory also includes the separation stage, whereby the mixture is depressurised, and products are separated by distillation into five product streams. The yield of each product has an estimated range depending on the exact conditions used in the reactor, and a value within that range has been taken for the purposes of the LCA study based on an average expected value per annum.

Table 1 – Yield of potential products from HTU

Product	Yield Range	Yield used for LCA
Process gas	10-15%	11.4%
Naphtha	20-30%	26.0%
Distillate gas oil (DGO)	20-30%	25.3%
Heavy gas oil (HGO)	20-30%	24.3%
Heavy wax residue (HWR)	10-20%	13.0%

The process itself differs from pyrolysis and other similar chemical recycling technologies by utilising supercritical conditions, meaning that the much of the thermal energy needed for the process can be provided by the process gas that is produced as one of the product streams.

The products are stored and the naphtha-equivalent fractions (naphtha & DGO, 51.3% of the total product) can be used as a steam cracker feedstock for the manufacture of polyolefins, replacing traditional petroleum naphtha are transported for upgrade at another vendor off site for bulk storage using a road tanker, prior to being shipped to the end user for use as a replacement for fossil naphtha as steam cracker feedstock, although the environmental burden of this process is not assessed as it is attributed to the downstream naphtha processing. All fractions from the HydroPRS process are registered as products under EU REACH (European Commission, 2006) and UK REACH (Department for Environment Food & Rural Affairs, 2021) and are therefore ‘lawful’, which is a criterion of the End of Waste under EU Waste Framework Directive (European Commission, 2008) and UK End of Waste criteria. Therefore, any further downstream burdens will be attributable to the onward processing by the product offtakers.

3. Results and Discussion

3.1. Feedstock Treatment and HTU Process Stages

The complete life cycle inventory of the HTU process and overall impacts generated through the LCA process can be found in the supplementary information. The overall Global Warming Potential (GWP) of the study, based on the currently available data, is 478 kg CO_2 eq. per tonne of input feedstock. The GWP of the two individual stages is given in Table 2. The results show that the contribution of the feedstock treatment is significant (28.6%) in the context of the overall process, highlighting the need to investigate the feedstock pre-treatment more thoroughly.

Table 2 – Breakdown of GWP contribution by stage

Stage	Contribution (%)
Feedstock treatment	28.6
HydroPRS process	71.4

3.2. Feedstock Treatment

Looking at the feedstock treatment in more detail (Table 3), the emissions due to electricity consumption has the largest contribution at 59% (80.2 kg CO_2eq). Waste processing activities have been assessed with the starting point feedstock treatment at the front end to protect the HTU process because feedstock is unlikely to be optimal. Carbon emissions as a result of the decomposition of the materials in landfill contribute 31% (42.2

kg CO_2eq). This would indicate that a simple way to improve the GWP of this section would be to improve the bale quality received by chemical recyclers with reduced levels of contaminants, particularly those that end up in landfill. Additionally, as this is an attributional model, no credits for the consequences of recycling the metals sifted out at this stage have been applied, as they would be attributed to the manufacturing of any subsequent product to include recycled metal content. Transport and other contributions are relatively small in comparison but could be mitigated through the use of electric vehicles to reduce the emissions generated.(Teelrn et al., 2021). Electricity consumption is the most significant contributor here, which will naturally decrease over time as the grid is made greener through planned improvements.(BEIS, 2020)

Table 3 – Detail of feedstock treatment contribution to GWP

Process	**GWP / kg CO_2eq**
Electricity consumption	80.2
Landfill	42.2
Transport & others	13.6
Total	136

3.3. Hydrothermal Upgrading

The GWP of the HTU process is estimated to be 341 kg CO_2eq. Examination of the relative contributions to GWP (Table 4) shows that the electricity consumption is the most significant contributor at 59% (201 kg CO_2eq). This is followed by process emissions at 37.2% (126kg CO_2eq), mainly reflecting process gas consumption in the high-pressure boiler but also consumption on-site in a low-pressure boiler for ancillary activities. The HTU process will reuse gas generated as a product stream so the process does not source natural gas from the grid, which is a benefit, but the gas is still being generated from what was originally a fossil-based source and the emissions should be accounted for. Other inputs into the process including water consumption and treatment are relatively minor.

Table 4 – Contribution of HTU process steps to GWP

Process	**GWP / kg CO_2eq**
Electricity consumption	201

Process emissions	126
Demineralised water consumption	13
Others	1
Total	341

3.4. Allocation and comparison to petrochemical production of naphtha

The HTU process produces five product streams, of which at least two (DGO and naphtha) can be combined and used as a replacement for fossil naphtha. This equates to 460 kg of naphtha-equivalent product being produced by 1 tonne of waste plastic processing. If the overall GWP of the process (478 kg CO_2eq) is allocated by mass to each individual product, that would mean 245 kg CO_2eq of the total GWP could be allocated to 460 kg of steam cracker feedstock production. As a comparison, the production of 460 kg of naphtha from crude oil at a refinery within the UK has a value of 184 kg CO_2eq. (Sphera Solutions, 2021a). If the feedstock processing stage was removed, or consolidated with other recycling activities such as mechanical recycling to produce a more optimal system, the value for the production of the steam cracker feedstock could be lowered as far as 175 kg CO_2eq. Additionally, the processes involved in the production of naphtha in a refinery are largely thermal processes that rely on energy to be produced using gas or the equivalent. There is also no accounting for flaring and other fugitive emissions in the quoted refinery model, and there is work that suggests that anthropogenic methane emissions have been underestimated (Hmiel et al., 2020). This suggests that the value of 184 kg CO_2eq is likely to be an underestimate of the real figure.

As HTU is more reliant on electrical energy, and with the grid expected to be made more carbon efficient over time, this transition will naturally reduce the GWP of HTU, to equivalent or better than fossil naphtha. Table 5 shows a sensitivity analysis for the electricity supply for the HTU process, looking at if the process was powered entirely by wind energy, or a prospective ‘net zero’ scenario based removing coal and gas from the current grid mix. There is a difference between the two prospective electricity scenarios because of the varying impact of renewable energy generation technologies such as solar, biomass etc. Furthermore, the modelling of HTU is of a prospective first-generation industrial process and it is highly likely that technical improvements made for future generations of HTU will reduce the environmental burden further.

Table 5 - Sensitivity analysis on electricity production

	Site grid mix	**100% wind power**	**"Net zero" scenario**
Climate change / kg CO_2eq	478	204	218

3.5. End of life treatment comparisons

Whilst naphtha production is an interesting comparator, it is not the main driver for the process as HTU is a waste treatment technology and is designed to deal with end-of-life plastic materials. Plastics will continue to be produced as they have key role within society, and as such, disposal measures need to be considered for their environmental metrics. Comparisons between climate change impacts for two end-of-life treatment technologies are shown in Figure 2. Secondary data for pyrolysis (Jeswani et al., 2021) and incineration (Sphera Solutions, 2021a) was used with the same scope and functional unit of processing one tonne of mixed plastic waste.

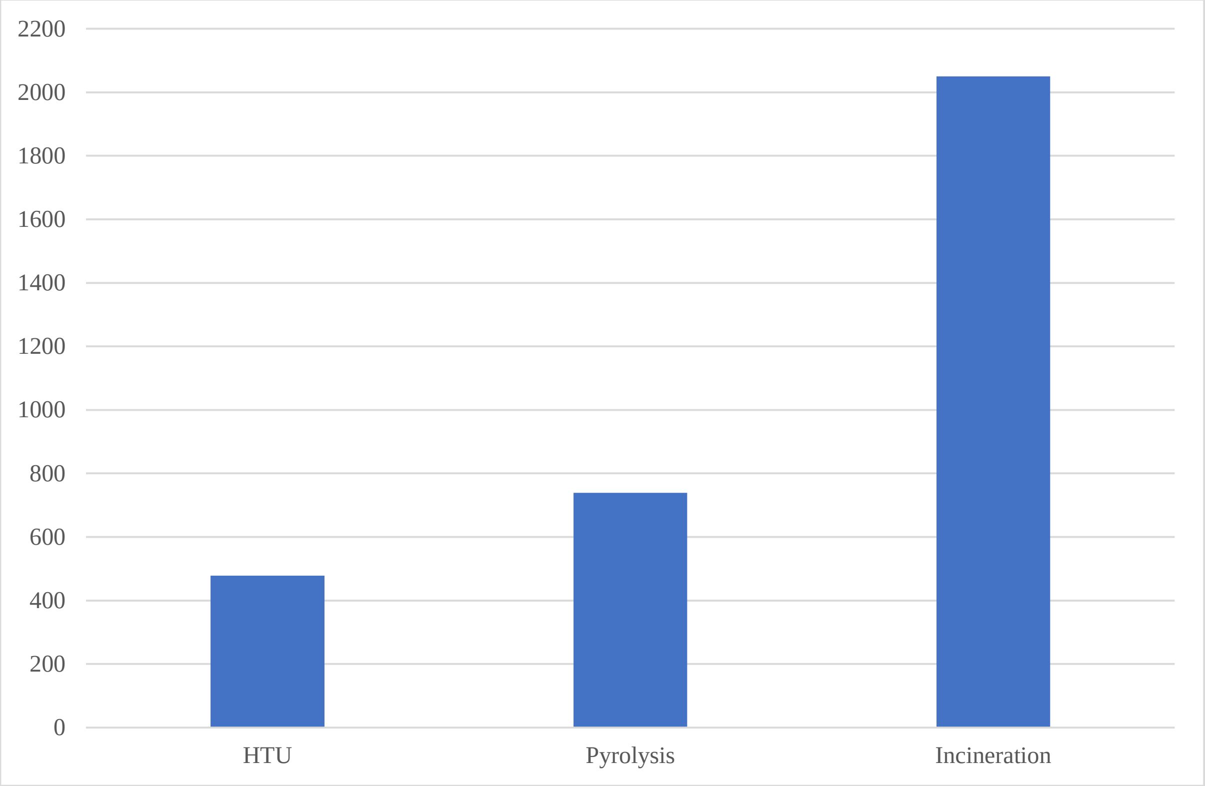

Figure 2 - Climate change impacts for end-of-life plastic treatment technologies

The results show that HTU has a 36% and 76% reduction in the climate change impact when compared to pyrolysis and incineration respectively. This is in line with another study commissioned by the Consumer

Goods Forum that has looked at the chemical recycling of polymers and shown similar levels of benefits when compared with incineration (Sphera Solutions, 2021b). Furthermore, incineration does not produce a product to retain in the system, so on top of the climate change impact of the disposal, there is a further cost of the production of virgin material to replace the plastic that has been lost from the system.

It could be argued that incineration has value because energy is generated from the system. However, incineration is an inefficient way (in terms of climate change impacts) of generating electricity as there are many other more climate-friendly options of generating electricity than waste incineration. When disposing of 895 kg of plastic *via* incineration, 5020 MJ of electricity is generated. Table 6 shows a comparison of the climate change impacts of generating 4990 MJ of electricity from a variety of conventional sources in the UK (Sphera Solutions, 2021a).

Table 6 – Climate change impacts of generating 5020 MJ of electricity

Electricity source	**Climate change / kg CO_2eq**
Hard coal	1370
Hydro	10
Incineration (plastic)	2050
Natural gas	610
Nuclear	7
Solar	94
Wind	8

It becomes apparent that incineration is significantly more damaging to the environment in terms of climate change than equivalent methods of generating electricity, including hard coal. This gives a further rationale for the introduction of new recycling technologies, and improvements to waste separation and sorting to remove residual plastic from refuse derived fuel.

4. Conclusions

The environmental impact of reprocessing hitherto unrecyclable waste plastic *via* a novel chemical recycling technology, HTU, has been assessed. The key driver behind the main impacts of this process is the consumption of electricity from the grid. This gives prioritisation for future research on the process to look at the reduction of electricity consumption through the implementation of energy efficiency technologies (e.g. heat exchangers) or looking more broadly from a national perspective at how electricity is generated, although this is an ongoing theme across governments and NGOs. The consumption of gas in the process could also be investigated – could steam generation be more efficiently achieved through electricity rather than gas? However, if that was employed, then a route for the utilisation of the gas generated by HTU would need to be investigated. This could potentially be achieved by condensing the gas into a liquefied natural gas which can be used as a steam cracker feedstock, a route commonly applied within the US. Furthermore, by removing the plastics from incineration, HTU offers a pathway to reduce carbon intensity of the grid by taking the incinerated plastic out of the system and having it replaced with low carbon electricity sources.

The impact in terms of climate change of naphtha produced via HTU is comparable to current fossil chemical feedstock production processes for future plastics (and other hydrocarbon products), presenting circularity options for plastics manufacturing alongside HTU as an end-of-life treatment option. Plastic will need to be disposed of and it is essential that this is conducted in the most environmentally safe way possible. HTU represents up to an 76% reduction in climate change emissions when compared with incineration, the main alternative method of disposal currently used for plastics that cannot be mechanically recycled. Furthermore, unlike incineration, both HTU and pyrolysis conserve the material within the system, leading to a reduction in consumption of fossil-based resources, with HTU operating at 36% less GWP than pyrolysis in the production of naphtha.

It has been considered that incineration has benefits in terms of energy generation. However, given that the incineration of waste plastics to produce electricity generates 50% more CO_2 per MJ than hard coal, it becomes clear that this should no longer be considered as an appropriate method of disposal for our waste materials as it is a sub-optimal method for both the treatment of waste plastics and the generation of electricity. As we move towards a more circular, net zero economy, these technologies should be phased out for more environmentally conscious routes. This therefore shows that the addition of chemical recycling to the end-of-life waste treatment options is essential within the global economy over the current fossil-based production and waste management system, from a combination of benefits in material circularity, reducing global warming potential and resource depletion standpoints.

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Synopsis

This paper presents an evaluation of environmental impacts of a novel chemical recycling technology for the processing of waste plastic.

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