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Technologies for chemical recycling of household plastics – A technical review and TRL assessment



Martyna Solis a,b,*, Semida Silveira

^a Energy and Climate Studies Division, KTH Royal Institute of Technology, Brinellvägen 68, Stockholm 10044, Sweden

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ABSTRACT

Chemical recycling is considered an attractive technological pathway for reducing waste and greenhouse gas emissions, as well as promoting circular economy. In the EU, readiness to develop a full commercial plant is becoming increasingly important given the ambitious goal to recycle all plastics by 2030. Household packaging streams tend to be of lower quality and lower recycling performance compared to industrial and commercial waste streams, thus requiring particular attention. This paper assesses chemical recycling technologies available and identifies the most suitable for recycling of household plastic waste. We identify eight different technologies and compare them in terms of process temperature, sensitivity to feedstock contamination and level of polymer breakdown, three critical factors affecting the cost and attractiveness of a chemical process. In addition, we carry out a Technology Readiness Level (TRL) assessment for eight technologies based on the stage of their present development. The review is based on peer-reviewed scientific papers and information collected from technology developers and providers, as well as interviews with experts. Our analysis outlines advantages and disadvantages of technologies available for chemical plastic recycling and their TRL. The chemical recycling technologies with the highest TRL are pyrolysis, catalytic cracking and conventional gasification. However, the economic feasibility of these technologies is difficult to assess due to the low number of projects in operation and scarcity of data available for comparison. The results of this analysis provide timely information as policy makers and developers set targets for recycling, and contemplate investments on research and chemical plastic recovering plants.

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Abbreviations: CEWEP, Confederation of European Waste-to-Energy Plants; FTI, Swedish packaging and newspaper collection; MSW, municipal solid waste; PET, polyethylene terephthalate; PS, polystyrene; PVC, polyvinyl chloride; PWMI, Plastic Waste Management Institute; RDF, refuse derived fuel; TRL, technology readiness level; WTE, waste-to-energy.

E-mail address: martyna.solis@profu.se (M. Solis).

^b Profu AB, Götaforsliden 13, 43134 Mölndal, Sweden

^{*} Corresponding author.

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1. Introduction

In 2015, the European Commission adopted the EU Plan for circular economy, paving the way for national strategies to address multiple challenges along the life cycle of materials such as plastics. The circular economy concept assumes a material production and consumption system in which waste material is recycled, recovered and reused, meaning the material is converted into a new product with economic value (Van Eygen et al., 2018). The main goals across Europe are to reduce the amount of plastic waste in circulation by enhancing material reuse, designing for recyclability and recycling with improved quality and economic gains (Ellen MacArthur Foundation, 2017). Plastic packaging is a priority as it accounts for approximately 60% of post-consumer plastic waste in the EU (European Commission, 2018).

In 2018, the EU Strategy for Plastics in the Circular Economy was adopted. The vision is that all plastic packaging shall be recyclable by 2030. Ambitious targets are set for the actual recycling of plastics. Common EU targets include 65% of recycled municipal solid waste (MSW) and 55% of plastic packaging recycled by 2035 (European Commission, 2018). It is estimated that 42% of plastic packaging was recycled in the EU in 2017, and between 14 and 18% at global level (Eurostat, 2019; OECD, 2018). According to the Confederation of European Waste-to-Energy Plants (CEWEP, 2019), 21% of European MSW was still being landfilled in 2016 as opposed to a target of maximum 10% by 2035. Conditions vary widely among EU member states, with less than 3% of MSW being sent to landfills in seven countries but more than half being landfilled in 11 countries.

Hestin et al. (2017) analyzed the main packaging resins from waste generated in five EU member countries: Germany, France, Italy, Spain and the UK. In all studied countries, household packaging streams tend to be of lower quality and lower recycling performance compared to industrial and commercial waste streams. Higher homogeneity and larger volumes of industrial waste contribute to higher recycling rates for post-industrial plastic waste in comparison with household waste. In this context, plastic waste recovery from households is an emerging key issue, requiring technologies that can help close the plastics loop, possibly producing new plastic products. Household waste plastic streams pose a challenge as they comprise a mix of contaminated plastic types with different recycling potential. As a result, chemical recycling is likely to play an important role in improving household plastic recycling rates in Europe.

Presently, a fraction of sorted plastic streams is recycled mechanically, leading to production of downgraded recycled material that is often more expensive than virgin plastics. Mechanical recycling cannot treat contaminated plastics or mixed plastic streams. Therefore, detailed and labour-intensive sorting is required before the recycling process. A significant amount of waste is thus rejected at an early stage, reducing the total efficiency of the process. Mechanical recycling results in approximately 10% material loss and 10% quality loss (Merrild et al., 2012). In addition, mechanical recycling degrades the material over its lifetime (Ragaert et al., 2017). According to the

Förpacknings och Tidnings Insamlingen (FTI, 2019), in the current system, plastic packaging can be recycled up to seven times before the polymers are too degraded to be used any further.

Chemical recycling provides an alternative to mechanical recycling, and could help to meet circular economy targets, while also resulting in recycled material of higher quality. Chemical processes have higher tolerance to mixed and contaminated plastic waste streams than mechanical recycling and can break down polymers into single monomers to produce a high-quality product. However, exactly how ready are chemical recycling technologies to help Europe and the world meet the challenges of plastic recycling?

The overarching objective of this study is to characterize and analyse chemical recycling technologies available for waste plastics as well as determine their current status and potential for future development. The comparison provides insight on key characteristics of chemical technologies that are suitable for recycling household plastic waste, and the level of development that they have reached. Knowledge about the technical maturity of available technologies is essential for investment decisions and implementation of solutions in commercial operations. The analysis is also relevant for setting targets and timetables or allocating resources for research and development to address household plastic recycling.

Following this introduction, Section 2 presents the methodology used to assess available technologies and parameters used to compare technologies. Section 3 describes the technologies analysed. Section 4 compares the technologies and determines their TRL. Section 5 concludes the paper.

2. Identifying and assessing technologies for chemical recycling

Four general categories of plastics recycling, and eight chemical recycling technologies were identified, which are considered in our assessment, see Fig. 1. The initial choice of technologies was done through a literature review. Chemolysis was excluded from the analysis because it is only suitable for treatment of homogenous plastic waste and we are dealing with heterogenous municipal plastic waste (Kumar et al., 2011; Ragaert et al., 2017). Thus, our analysis focuses only on cracking and gasification technologies, see Fig. 2. The technologies are assessed with focus on key performance parameters. The assessment relies on peer-reviewed scientific papers as well as information provided by technology developers via their respective websites. The findings are complemented with expert interviews.¹

Each technology is evaluated and compared based on three key parameters: process temperature, sensitivity to feedstock contamination and level of polymer breakdown. Given the small number of large-scale projects in operation, real technical data is scarce. This makes it challenging to compare the technologies. Our paper focuses on comparing the technologies based on publicly available data.

¹ Tobias Richards, Professor in Resource Recovery and Building Technology at University of Borås; Henrik Thunman, Professor at Department of Energy and Environment at Chalmers University of Technology

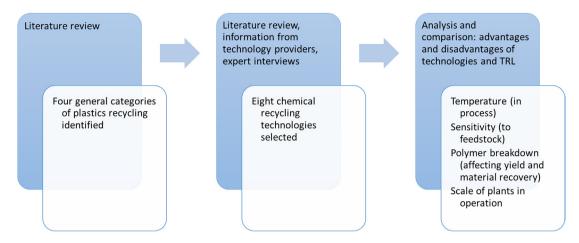


Fig. 1. Definition of suitable chemical technologies for household waste recycling, and parameters compared.

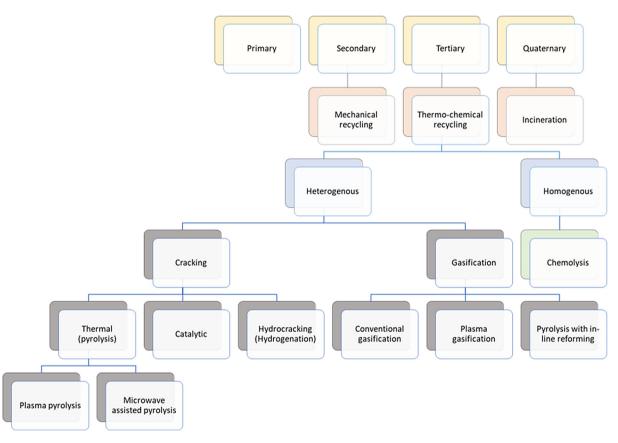


Fig. 2. Routes for recycling of solid plastic waste, based onKumar et al. (2011). Note: Chemical recycling technologies for treating plastics in household waste streams are shown in grey.

Table 1Framework for assessing the Technology Readiness Level (TRL) of technologies available for plastic recycling. *Source*: Adapted from Thunman (2018).

Technology developer	Comment	TRL
Large sized company	In development/in commercial operation	8-9
Small/medium sized company	Successfully sold/in commercial operation	8–9
Small/medium sized company R&D centre/university	In development	4–7 1–5

Higher temperature allows more comprehensive polymer breakdown and higher material purity of the processed material. This provides higher material recovery potential and enhanced flexibility for the portfolio of products that can be generated (Balakrishnan and Sreekala, 2016). The temperature is particularly important when treating mixed waste plastic. Different polymers have different melting temperatures and will need slightly different conditions to break down into monomers. The suggested process temperature is the temperature suitable for the highest melting component, though this may potentially lead to overheating and degradation of lower melting components (Ragaert et al.,

2017). High operating temperatures result in higher costs and required energy input.

The lower the process temperature of a technology, the more sensitive it is to the quality of the waste. With low temperatures more detailed waste separation is required before chemical recycling. With advanced separation techniques available today, additional waste separation is not considered an obstacle to full-scale implementation of chemical recycling technologies with lower process temperature. However, this may result in additional costs other than recovery plant costs, and will require more effort around logistics.

The sensitivity of different technologies to waste contamination determines the extent of efforts needed around logistics. The more sensitive the technology, the more detailed waste separation is required. The sensitivity of a technology can be reduced by increasing process temperature.

If high polymer breakdown can be achieved, the technology is suitable for new plastics production. This can reduce demand for virgin plastics and contribute to closing the plastics loop. A more detailed breakdown of plastic input implies less need for upgrading steps before new plastics can be produced. The possible polymer breakdown is, thus, crucial when assessing the potential contribution of a technology to achieving circular economy targets.

The Technology Readiness Level (TRL) assessment is carried out based on the comparison of the type and size of the entity that is developing the technology, see Table 1. Technologies that have only been developed at academic or research level have a low TRL. Medium to high range TRL refers to small and medium sized companies with prototype or ready technology, which can be successfully scaled up. Technologies developed by large companies with significant R&D resources and opportunities to explore economies of scale have the highest TRL. The latter are the most likely to succeed in the near future. The TRL levels are assigned to respective developers according to the TRL scale given by (Rybicka et al., 2016).

3. Technologies available for recycling of plastics and their characteristics

There are four general categories of plastics recycling, see Fig. 2 (Singh et al., 2017):

Primary recycling, so-called closed loop recycling, is mechanical reprocessing of single uncontaminated plastic, resulting in a product with equivalent quality;

Secondary recycling is mechanical recycling that downgrades the recycled material;

Tertiary recycling is chemical recycling or feedstock recycling, which breaks down polymers to monomers with quality recovery of the recycled material;

Quaternary recycling is waste incineration for energy recovery. Primary recycling is not suitable for municipal solid waste (MSW) because it cannot process contaminated mixed plastics. Secondary recycling requires sorting which generates high material losses. Tertiary recycling offers recovery of material quality, leads to raw material production, supports the reuse of plastics and is well in line with current ambition to close the loop on plastics. Plastic waste offers high potential as a source for chemicals and fuels production (Achilias et al., 2007; Singh et al., 2017). Finally, the quaternary method shortens the material's lifespan. It is used in many countries but is, in many ways, an interim solution as it is not fully aligned with the evolving principles of a circular economy.

This paper focuses exclusively on the tertiary (chemical) recycling technologies, which are the ones considered to be a potential solution to recycling of plastics from household waste – cracking

and gasification technologies, see Fig. 2. Chemical recycling are processes in which a plastic polymer is chemically broken down to monomers and subsequently restructured into new polymers used for new plastic production. This paper does not consider any other type of recycling, for example bio recycling.

3.1. Thermal cracking (conventional pyrolysis)

Conventional pyrolysis is a suitable technology for waste plastics that are difficult to depolymerize, such as multilayered plastic packaging. These are currently incinerated. The process takes place at moderate to high temperatures, and in absence of oxygen. The key difficulties are the complexity of reactions and the high amount of energy required in the process (Anuar Sharuddin et al., 2016).

The process may be challenging when the plastics are mixed with contaminants (Ludlow-Palafox and Chase, 2001). Pyrolysis has low tolerance to the PVC present in the feedstock. Then chlorinate compounds can be formed in the pyrolysis oil which makes its use difficult (López et al., 2011).

Thermal cracking is a simple technology, in which the process parameters can be changed to optimize the product yield according to preferences and needs. Parameters that affect the final products are temperature, pressure, residence time, introduction of catalysts and heating rate (Gabbar et al., 2017). The products from waste plastics thermal cracking are gas, char and liquid oil. In many cases pyrolytic oil is the most desired product from the process due to its relatively high calorific value and many potential applications, for example in petroleum blends. Pyrolytic oil often requires upgrading before further use. The process temperature ranges between 300 and 700 °C (Anuar Sharuddin et al., 2016; Das and Tiwari, 2018).

Examples of operational full-scale plastic pyrolysis are two plants located in Sevilla and Almeria, Spain, owned by Plastic Energy. The company uses thermal anaerobic conversion which involves melting and gasification of plastics followed by condensation and refining (Plastic Energy, 2019a). The plants can process all types of plastic waste. However, the process efficiency and profitability vary depending on feedstock mix and quality. The products from the process are alternative fuels or oil used for new plastics production. The company has recently announced that it will build five plants in Indonesia (Plastic Energy, 2019b). The list of industrial facilities working with plastic thermal cracking is presented in Appendix A. In recent years two pyrolysis technologies have been gaining attention: plasma pyrolysis and microwave-assisted pyrolysis.

3.1.1. Plasma pyrolysis

Plasma pyrolysis integrates the conventional pyrolysis with thermochemical properties of plasma to transform plastic waste into syngas. The process temperatures are very high, ranging between 1730 and 9730 °C, and the waste plastics are decomposed into monomers. The process is extremely fast, lasting between 0.01 and 0.5 sec, depending on process temperature and type of waste (Dave and Joshi, 2010). The resulting syngas is composed mainly of CO, $\rm H_2$ and small amounts of higher hydrocarbons.

Plasma pyrolysis has several advantages over conventional pyrolysis, for example, the comprehensive polymer breakdown. The produced gas has a low tar content and high heating value which makes it suitable for electricity generation in turbines or hydrogen production. Plasma pyrolysis is an appropriate method for mixed waste plastic feedstock and a promising technology for gaseous fuel or chemical production purposes. It solves the problem of toxic compounds in syngas as the temperature is high enough to decompose them and limit the formation of free chlorine from HCl, which is a common problem in other chemical

recycling technologies. Furthermore, the technology has significantly low emissions levels (Punčochář et al., 2012).

Thermal plasma technology is a well-established technology in metallurgy processing or material synthesis (Dave and Joshi, 2010). For now, due to economic and legal aspects, the most important application of the technology is the destruction of hazardous waste rather than recycling (Moustakas et al., 2005). Plasma pyrolysis of waste plastics has been investigated only at laboratory scale, see Appendix B (Gabbar et al., 2017; Guddeti et al., 2000; Tang et al., 2013). There are still several technical challenges to be addressed before the technology can meet waste management requirements and become commercially available. The challenges differ depending on the specific plasma technology. Tang et al. (2013) indicate that there is great interest in finding a suitable plasma technology for waste treatment application and conclude that spout-fluid bed reactor technology may be an interesting solution.

3.1.2. Microwave-assisted pyrolysis

Microwave-assisted pyrolysis of waste plastics involves mixing plastics with a highly microwave-absorbent dielectric material. The heat absorbed from the microwaves is transferred to the plastics by conduction. The sources of microwave radiation allow very high temperatures and heating rates and reach high conversion efficiencies of electrical energy into heat and heat transfer to load (Arshad et al., 2017; Ludlow-Palafox and Chase, 2001).

The process has several advantages over a conventional process. It offers even heat distribution, higher heating rates, more control over the process and increased production speed when compared to the conventional pyrolysis. The technology is also characterised by selective material heating. The magnitude of heating depends on the dielectric strength of the material. In general, plastics have poor dielectric strength and, when mixed with dielectric absorbent, the heating efficiency may vary for different materials. Large fluctuations in plastic waste composition could be a challenge. Since heating efficiency varies for different absorbents, it may be difficult to use dielectric absorbents efficiently on industrial scale. Another challenge is insufficient information to quantify the properties of dielectric materials which determine the heating efficiency in the process (Arshad et al., 2017).

Ludlow-Palafox and Chase (2001) tested the potential of microwave-assisted pyrolysis for recycling of packaging, including aluminium and polymer laminates often present in MSW. The results showed that the technology is suitable to treat these materials and could possibly serve to treat real waste.

So far, the microwave-assisted pyrolysis has been studied at laboratory (Aishwarya and Sindhu, 2016) and pilot scales (Akesson et al., 2013). Between 2003 and 2005, Stena Metall together with several other partners established a project to explore the potential of microwave-assisted pyrolysis as an alternative method for recycling glass fibre reinforced composites. Both laboratory scale equipment and pilot plant were developed. The results showed that the technology requires more development and large production volumes to become commercially feasible (Stena Metall, 2012). The technology has not been analyzed at industrial scale partly due to insufficient data to quantify the dielectric properties of treated waste streams, which is needed for the estimation of microwave heating efficiency (Anuar Sharuddin et al., 2016).

3.2. Catalytic cracking

Adding a catalyst to the pyrolysis process may help save energy and reduce production costs since catalysts reduce the required process temperature. With a catalyst added, the process temperature can be lowered to 300–350 °C as opposed to around 450 °C for pyrolysis. Introducing a catalyst can also increase the yield of

products with higher added value (Almeida and De Fátima Marques, 2016). Catalysts have been used for product upgrading and for obtaining oil with properties similar to those of fossil fuels (Anuar Sharuddin et al., 2016). Most of the work on plastic catalytic cracking has been performed with pure polymers since the process may be affected by contaminants present in the mixed waste plastic stream (Achilias et al., 2007; Almeida and De Fátima Marques, 2016).

Catalytic cracking has lower operating temperature and higher oil yield than the conventional cracking process for most plastics, if the right catalyst is selected. Catalysts speed up the reaction and remain unchanged by the process (Aguado and Serrano, 1999). Conversion as high as 100% is achievable in plastic waste cracking, and the oil yield ranges between 86 and 92% (Sahu et al., 2014). The main difficulty is that the chloride and nitrogen components present in the raw waste stream tend to deactivate the catalyst, whereas the inorganic materials tend to block the catalyst's pores. Therefore, pre-treatment of the waste is often required as a preventive action (Ragaert et al., 2017).

Catalysts are used to optimize product distribution and selectivity, especially in the context of automotive fuels and chemicals used in the petrochemical industry (Miandad et al., 2017). Catalytic decomposition of polymers follows the same reaction steps as hydrocarbon catalytic cracking used in petroleum refineries, and the catalysts used are also similar.

There are several commercial catalytic cracking processes at industrial scale. An overview is provided in Appendix C. Multiple companies all over the world work with the technology (Samperio, 2016). One of world's largest catalytic cracking projects was Sapporo Plastics Recycling which, together with Toshiba, coowned the world's largest waste plastic liquefaction facility in Japan. The facility converted 15,000 tonnes of mixed waste plastic into light oil, which was used as feedstock for new plastic products, medium fuel oil equivalent to diesel and heavy oil used for electricity generation. However, Sapporo Plastic Recycling withdrew from the business in 2010 due to financial problems (Fukushima et al., 2009; Klean Industries, 2015; PWMI, 2016).

3.3. Hydrocracking

Hydrocracking involves adding hydrogen to the cracking process which results in higher product quality. The process occurs at elevated hydrogen pressures, approximately 70 atm, and temperature range from 375 to 500 °C (Ragaert et al., 2017). The waste plastic is subjected to low temperature pyrolysis before the main process leading to plastic liquefaction freed from non-distillable matter. The liquid is then sent over to a catalyst bed. A catalyst plays an important role in the hydrocracking process since it reduces the reaction temperature and increases the oil yield and quality. However, Munir et al. (2017) found that the presence and type of catalyst does not have significant influence on the process at elevated temperatures. The biggest obstacle in implementing this technology is the cost of hydrogen. Electrically produced hydrogen can cost roughly €2500 per tonne (Ragaert et al., 2017).

Hydrocracking technology is present in other industries, for example petroleum industry (Ding et al., 1997). Although it has been tested for waste plastics feedstock, it is only available at pilot scale for this purpose, see Appendix D. Several challenges remain before it is commercially viable, for instance poisoning effect after hydrocracking of PVC (Richards, 2018). High investment and operating costs will have to be optimized.

3.4. Conventional gasification

Gasification of waste plastics results in a mixture of hydrocarbons and synthesis gas (syngas), with emphasis on maximizing the yield of the latter. Gasification of plastic waste can be used to produce energy, energy carriers such as hydrogen, as well as chemicals from syngas. The solid waste gasification usually occurs at temperatures between 700 and 1200 °C depending on the gasifying agent; the main ones are air, steam and plasma. The gasifying agent determines the composition of the syngas produced and its applications. The gas in the waste plastics gasification has higher tar content than gases produced from biomass or coal gasification, and thus the overall process efficiency is reduced (Devi et al., 2003; Lopez et al., 2018). The technical challenge is to improve the quality of the syngas so that it is suitable for different applications (Arena et al., 2010).

During gasification two undesirable by-products, tar and char, are produced. The actual amounts depend on the gasified plastic waste characteristics. Since most commonly used polymers like PS. PVC or PET have a high volatile content and can be almost entirely converted into volatiles under fast pyrolysis conditions. the char yield is very low (Anuar Sharuddin et al., 2016). Higher amounts of char production occur only when plastic waste contains other materials such as biomass and fibers (Lopez et al., 2018). On the other hand, high volatile content of plastics results in higher tar formation (Mastellone et al., 2010; Pinto et al., 2009). The air gasification results in syngas diluted by atmospheric nitrogen, up to 60%, which has too low calorific value for use in a gas turbine (Lopez et al., 2018). The new generation turbines, however, can efficiently combust low heating value syngas, if it is cleaned and partially cooled. The oxygen-enriched air gasification, with oxygen content between 21 and 50%, results in syngas with higher heating value which could serve as feedstock for processes at higher temperatures. The pure oxygen gasification generates syngas almost free from atmospheric nitrogen, thus with higher calorific value. This type of gasification process requires oxygen separation from air, which is carried out in an air separation unit. This is very cost intensive and only viable for large-scale units above 100,000 tonnes of feedstock per year (Arena, 2012). The steam gasification results in nitrogen-free syngas with high heating value and high hydrogen concentration. It can be used for synthesis applications and production of new plastic products.

An operational full-scale plant based on gasification technology with potential to contribute to a circular economy is owned by Enerkem, and located in Edmonton, Canada. It converts 100,000 tonnes of dried and post-sorted waste annually into 38 million litres of biofuels: methanol, then ethanol and ethylene. The plant uses steam gasification technology in a fluidized bed reactor. The conversion process lasts less than ten seconds and, afterwards, the syngas is cleaned and conditioned. Enerkem's technology breaks all the plastics down to hydrogen and carbon monoxide, and produce biofuels from it. The process requires high level of waste separation and very large amounts of energy (Chemicals Technology, 2019; Nistelrooij, 2017). The company is part of a consortium planning a waste-to-chemicals plant in Rotterdam, the Netherlands. The new plant shall have capacity to convert up to 360,000 tonnes of waste into 220,000 tonnes (270 million litres) of methanol (Enerkem, 2018).

The industrial facilities based on plastic gasification process are listed in Appendix E.

3.5. Plasma gasification

Plasma technologies have been used for more than 30 years in different applications, such as chemical, metal and space industries. Plasma technology is mainly used to decompose hazardous waste, and its introduction to waste-to-energy (WTE) industry is relatively new (360 Recycling, 2019).

Plasma gasification is a process in which plasma torches create electric arc via passage of electric current through a gas. The process temperature is very high, up to 15,000 °C, and can be controlled independently from fluctuations in feedstock properties and quality. Plasma gasifiers have high tolerance to low quality feedstock (Arena, 2012; Gomez et al., 2009).

Plasma gasification enhances conventional gasification process and results in higher purity of product gas with reduced level of tars. Plasma, an electrically conducting medium, is generated through plasma torches by heating up gas (usually air) to extremely high temperatures, up to approximately 3900 °C. As a result, the gas ionizes and creates plasma (Pourali, 2010). Untreated waste comes in contact with electrically generated plasma in the reactor. Process conditions are usually atmospheric pressure, and temperature usually ranges between 1500 and 5000 °C but can reach up to 15,000 °C. The residence time is usually very short, in the order of minutes. Organic matter in the feedstock is converted into syngas whereas inorganic matter is turned into inert slag.

Plasma gasifiers have a very high electricity requirement to operate. For a plasma gasification plant with around 1200–2500 MJ/ton of waste, the electricity requirement would be 5–10% of the energy in the waste and 15–20% of the gross power output (Arena, 2012; Lombardi et al., 2012). Increased electricity requirements and improved gas quality imply higher operating costs and larger investments.

Some of the first commercial applications of plasma gasification are located in Japan - a WTE plant owned by Westinghouse Plasma Corporation and Hitachi Metals located in Eco Valley and a WTE plant owned by Hitachi Metals located between cities of Mihama and Mikata (Willis et al., 2010). Other operating plasma gasification plants are located in China and India (Alter NRG, 2019; Westinghouse Plasma Corporation, 2014). Plasma gasification is not available in Europe. There were plans to construct two plasma gasification plants in Tees Valley, UK, to process 2000 tonnes of RDF per day. In 2016, Air Products, the project developer had to close down due to technical difficulties in plant operations. The investment cost made the project economically unfeasible (Messenger, 2016).

3.6. Pyrolysis with in-line reforming

The increased interest in pyrolysis reforming process lies in high hydrogen production from the process, which usually exceeds 30%, and the gas is free of tars. The process temperature varies between 500 and 900 °C depending on the feedstock, reactor configuration and bed material (Lopez et al., 2018). The process is carried out in two connected in-line reactors for pyrolysis and reforming steps. The impurities in the plastic waste remain in the reactor and they do not have any contact with the reforming catalyst. This prevents the catalyst from deactivating. The process temperature is lower than in gasification which lowers the production costs. The tars from the gas product can be fully eliminated. The hydrogen production obtained is significantly higher than in the steam gasification process.

Despite great potential, the studies of the technology are currently limited to laboratory scale (Barbarias et al., 2016; Erkiaga et al., 2015; Lopez et al., 2018). An experiment carried out by (Wu and Williams, 2010) showed that lower gas yield was obtained from the non-catalytic process, while adding catalyst to the process enhanced the hydrogen production. Ouadi et al. (2017) developed a 2 kg/h pilot plant for pyrolysis and in-line reforming of MSW. Before the technology can be implemented at industrial scale several challenges need to be addressed, for instance, detailed investigation of the catalyst deactivation (Lopez et al., 2018). A study by (Barbarias et al., 2018) proved the feasibility of fast pyrolysis and in-line reforming of different plastics and plastic mixture. The process was performed in a

Table 2Advantages and disadvantages of cracking and gasification technologies analysed in the study.

Technology	Advantages	Disadvantages	Example of available installations
Thermal cracking (conventional pyrolysis)	Simple technology Flexible process Suitable to waste plastics that are difficult to depolymerize	Complexity of reactions High energy requirement Low tolerance to PVC Sensitive to contamination of the feedstock Products often need upgrading before further use	Mogami-Kiko, Japan, 3 tonnes/day, commercial scale operation
Plasma pyrolysis	High enough temperature to decompose the toxic compounds in product gas – suitable for mixed plastic waste Product gas with low tar content and high heating value Limited formation of free chlorine from HCl	High electricity requirement	Laboratory scale operation
Microwave assisted pyrolysis	Even heat distribution Suitable to treat MSW In comparison to conventional pyrolysis: higher heating rates more control over the process increased production speed	Sensitive to large fluctuations in waste composition Challenge of efficient use of dielectric absorbents on industrial scale Requires large feedstock volumes to be feasible	Laboratory and pilot scale operation
Catalytic cracking	In comparison to thermal cracking: lower operating temperature higher oil yield shorter reaction time possible reduced production cost possible reduced energy consumption Product with similar properties to fossil fuels Catalyst helps optimize product distribution and selectivity Possible 100% conversion for plastic waste	Sensitive to contamination of the feedstock Chloride and nitrogen components in waste can deactivate the catalyst Organic material in waste can block catalyst's pores Often requires pretreatment	Sapporo/Toshiba, Japan 14.8 ktonnes mixed waste plastic/year, commercial scale operation
Hydrocracking	High quality product	High cost of hydrogen Poisoning effect after hydrocracking of PVC	Pilot scale operation
Conventional gasification	Possible very detailed polymer breakdown: to hydrogen and methane Multiple applications of the product gas Suitable for mixed plastic waste Well-established technology The pure oxygen gasification generates syngas almost free from atmospheric nitrogen The steam gasification results in syngas free from nitrogen which can be used for synthesis applications and production of new plastic products Possible hydrogen production from steam gasification	High investment and operating costs The product gas requires improving the quality before it is further used Requires high feedstock volumes to be feasible Tars and char in product gas Cost and energy intensive Pure oxygen gasification requires oxygen separation from air, which is very cost intensive	Enerkem, Edmonton, Canada, 100 ktonnes RDF/year, commercial scale operation
Plasma gasification	In comparison to conventional gasification: higher purity of product gas reduced level of tars High tolerance to low quality feedstock Temperature control independently from fluctuations in feedstock properties and quality and gasifying agent supply Well-established technology	High investment and operating costs Very high electricity requirement	No commercial operation in WTE industry, widely used to decompose hazardous waste on commercial scale
Pyrolysis with in-line reforming	Hydrogen production from the process Product gas free of tars The impurities in the plastic waste do not have any contact with reforming catalyst In comparison to conventional gasification: lower process temperature lower production cost higher hydrogen production than from steam gasification	Catalyst deactivation issue needs to be further researched and solved	Pilot scale operation

continuous regime which is an encouraging result and a milestone for development of this technology.

4. Comparing technologies for chemical recycling and determining their TRL $\,$

Table 2 summarizes advantages and disadvantages of the cracking and gasification technologies for plastic recycling described in Section 3.

Conventional pyrolysis operates at the lowest temperature range if compared to the other evaluated technologies. Sensitivity to feedstock contamination is high and, if the waste contains PVC, chlorinate compounds can be formed in the pyrolysis oil, making

its use difficult. Pyrolysis results in moderate breakdown of molecules and relatively low quality of the final product, which would require upgrading before further use. According to Thunman (2018), due to the chemical properties of different components in the pyrolytic oil, only a small portion can be used for new plastic production. Thus, the potential to obtain plastics of virgin quality through this process will be low.

Microwave-assisted pyrolysis operates at higher temperatures than conventional pyrolysis and allows more detailed polymer breakdown. The technology is sensitive to large fluctuations in the plastic waste composition, as mixing dielectric absorbent with different plastic materials may result in different heating efficiency, and affect the ratio of heat output to input. This affects

Table 3Key parameters of analysis in technologies for chemical plastic recovery.

Technology	Scale of operation (at present)	Temperature (°C) (in process)	Sensitivity (to feedstock quality)	Polymer breakdown (affecting yield and material recovery)	TRL
Conventional pyrolysis	Commercial	300-700	High	Moderate	9
Plasma pyrolysis	Laboratory	1800-10000	Low	Very Detailed	4
Microwave assisted pyrolysis	Laboratory	Up to 1000	Medium	Detailed	4
Catalytic cracking	Commercial	450-550	High	Moderate	9
Hydrocracking	Pilot	375-500	High	Detailed	7
Conventional gasification	Commercial	700–1200	Medium	Detailed	9
Plasma gasification	Commercial in decomposing hazardous waste	1200-15000	Low	Very detailed	8
Pyrolysis with in-line reforming	Pilot	500–900	Medium	Detailed	4

the speed of the process for different materials and makes the process challenging.

Catalytic cracking allows process design and temperature control for obtaining the desired end-product. The process temperature is comparable to the process temperature of thermal cracking. The technology is highly sensitive to feedstock contamination as chloride and nitrogen in waste can deactivate the catalyst, and the inorganic materials block the catalysts' pores. The use of catalyst allows moderate polymer breakdown while lowering temperature requirements. Just like in the case of thermal cracking, the final product needs upgrading before further use.

Hydrocracking operates at low temperatures. The use of hydrogen in the process allows detailed polymer breakdown. The technology cannot be used for PVC due to the resulting poisoning effect.

Conventional gasification operates at higher temperature and offers more detailed breakdown than thermal and catalytic cracking. The gasification process can support the reuse of plastic material and has been used to produce intermediate chemicals and various plastics, for example in the Enerkem plastic waste to methanol/ethanol facility in Canada. When driven to its extreme, the process can convert all organics in the plastics into hydrogen and carbon monoxide (Thunman, 2018). The resulting product has high purity with multiple possible applications. During gasification, undesirable products, such as tar and char, are produced in amounts determined by the content of the feedstock. Most plastic polymers have high volatile content, and their gasification will result in tar and char formation. The quality of the gas needs to be improved before it can be further used.

Plasma technologies operate at the highest temperatures among all considered technologies and can serve to obtain the most detailed molecules breakdown, hydrogen and carbon monoxide. These technologies are expected to provide the highest level of quality recovery of processed waste plastics. Most likely, the product obtained does not need upgrading before use, but the high energy input can make the process unfeasible.

Pyrolysis with in-line reforming operates at temperatures lower than those required in conventional gasification, and allows detailed polymer breakdown. The catalyst does not have direct contact with any impurities in the feedstock, which prevents it from deactivating. This means less sensitivity to feedstock characteristics than in catalytic cracking or hydrocracking.

Table 3 summarizes the three key parameters analyzed in the different technologies, namely process temperature, sensitivity to waste contamination and contribution to material recovery. The material recovery potentials of the technologies should be considered as preliminary.

Conventional pyrolysis, catalytic cracking and hydrocracking operate at the lowest process temperatures, and show highest sensitivity to feedstock characteristics. Conventional pyrolysis and

catalytic cracking offer moderate polymer breakdown. Polymer breakdown in hydrocracking is detailed as a result of the use of hydrogen in the process.

Microwave-assisted pyrolysis, conventional gasification and pyrolysis with in-line reforming operate at higher process temperatures and medium sensitivity to feedstock contamination. They offer detailed polymer breakdown.

Plasma technologies, plasma pyrolysis and plasma gasification operate at the highest process temperatures and lowest sensitivity to feedstock contamination. They offer very detailed polymer breakdown.

Hydrocracking, microwave-assisted pyrolysis and pyrolysis with in-line reforming have not been tested at industrial scale for waste management purposes, and further research is required before these technologies can be attractive.

With the information gathered, we have determined the TRL for the different technologies. The commercially available technologies, with a TRL 9, are pyrolysis, catalytic cracking and conventional gasification. The current plasma gasification projects use the technology for post-treatment, that is, for treating hazardous waste as well as solid residues from the gasification process rather than for processing the main solid residue itself. Therefore, plasma gasification for plastic waste is considered to have TRL 8. The technology has potential to soon be commercially available (Richards, 2018). Hydrocracking has TRL 7 as the largest obstacle in implementing the technology at industrial scale is the cost of hydrogen. The technology is available at pilot scale and developed by large sized companies. Plasma pyrolysis, microwave-assisted pyrolysis and pyrolysis with in-line reforming were classified at TRL 4. They are available at laboratory scale, and further research is required before they can be commercially available.

5. Conclusions

To achieve its ambitious target on circularity and plastic recovery, the EU will have to explore new technological options. Chemical recycling could be part of the solution to increase recycling of household plastic waste. This study assessed eight chemical recycling technologies for dealing with mixed waste plastics from households and determined their TRL. The results indicate that there are three technologies with TRL 9: thermal cracking (pyrolysis), catalytic cracking and conventional gasification. Other technologies are promising but require more effort as they are not at commercial stage yet. In general, the higher the temperature, the higher the expected purity of final output products, and potential for monomers recovery from the process. However, that comes at a cost in terms of higher energy requirements. Ultimately, technology options for waste recovery will have to be aligned with sustainability goals and the overall objective to transition towards a circular economy.

At present, there is limited data and few full-scale projects in operation to support a solid economic feasibility analysis of the different technologies for chemical recycling. Yet, the study shows that there are mature technologies that can be used at large scale to help Europe reach its plastic recycling targets. The choice of technology and its cost-efficiency in a specific case will depend on factors such as available feedstock volume, content of the feedstock, local policy as well as gate fees charged for MSW recycling.

The results from this comparative study provides a starting point for potential investors or institutions aiming at technological solutions for the plastic waste problem. The results serve also to signal to policy makers what opportunities are at hand and indicate potential areas of focus for increased research and development. However, it is important to remember that chemical technologies are only one part of the solution for plastic recovery. Other measures include, for example plastic design, reduction in the use of plastics and plastic waste separation to obtain more homogeneous feedstock. With better information on the characteristics of the plastic waste and its recyclability, more choices for material conversion will be available. This would affect the choice and cost of the chemical recovery processes as well.

Appendix A

See Tables 4 and 5.

Table 4 Industrial facilities for plastic pyrolysis. *Source*: Adapted from Butler et al. (2011), Plastic Energy (2019), Ragaert et al. (2017) and Samperio (2016).

Process	Temperature, °C	Feedstock, %	Yield
PYROPLEQ	450-500	PW, MSW	Energy
Akzo Nobel	700-900	PVC rich PW	HCl, CO, H ₂ , CH ₄
PKA-Kiener	450-500	PW, MSW	Gas, energy
Siemens - KWU	450-500	PE, PP, PS	65% gas, 35%
			solid residue
DBA process	450-500	PW	Energy
Kobe Steel	450-500	PW	Oil and gas
Ebara	Unknown	PW	Energy
Mogami-Kiko	Unknown	PP (67), PE (33)	79% oil, 12% gas
Hitacho-Zosen	Unknown	PE (55), PP (28),	84% oil, 10% gas,
		PS (17)	6% solid residue
Royco Beijing	Unknown	PE, PP, PS,	87% oil, 10% gas,
		waste oils	3% solid residue
Chiyoda process	Unknown	PW	50% oil, 16% gas,
			34% solid residue
Plastic Energy	Unknown	Plastic waste	Oil, alternative fuels

Table 5Capacities of some of the commercial or pilot plants working with plastic pyrolysis processes. *Source*: Ragaert et al. (2017).

Process	Location	Capacity	Status
Mogami-Kiko	Japan	3 t/d	Operational
Royco Beijing	China	6 kt/d	Unknown
Sapporo/Toshiba	Japan	14.8 kt/y	Operational
ALTIS	Japan	unknown	Commercially applied
Gossler Evitec	Germany	1 kt/y	Unknown
Changing World Technologies	USA	31,470 toe/y	Demo

Appendix B

See Table 6.

Table 6Different plasma pyrolysis/gasification reactor systems for waste treatment application. *Source*: Adapted from Tang et al. (2013).

Reactor	Plasma source	Input/output
Plasma moving bed reactors	DC	MSW/hydrocarbons
Plasma fixed bed reactors	DC, air, 200 kW	SW/hydrocarbons
Plasma fixed bed reactors	DC – RF, argon/oxygen, 34 kW	Charcoal deformed after treatment

Appendix C

See Table 7.

Table 7Industrial facilities for plastic catalytic cracking.Klean *Source:* Adapted from Butler et al. (2011), Fukushima et al. (2009), Industries (2015), Ragaert et al. (2017) and Samperio (2016).

Process	Temperature, °C	Catalyst	Feedstock	Yield
Zadgaonkar	350	Unknown	PE, PP, PS, PVC, PET	75% oil, 20% gas, 5% coke
Smuda	350	Ni-silicate, Fe-silicate	PE, PP, PS, PVC, PET	95% oil
T-technology	390-420	Unknown	PE, PP, PS	78% oil
Fuji	390	HZSM5	PE, PP, PS	75% oil
Amoco	490-580	Unknown	PE, PP, PS	oil, gas
Mazda	200-450	Al ₂ O ₃ , ZrCl ₄	ASR, PE, PP, PS, PU, ABS	60% oil
Nikko	200–250	Metal catalyst	PW	80% oil
Reentech	350–400	Al-silicate	PE, PP, PS, PVC	75% oil, 15% gas, 10% solid residue
NanoFuel	270-370	HY	PP, PE, biomass	94% oil
Thermofuel/ Cynar	350-425	Metal catalyst	PE, PP, PS	Oil, gas, coke
Fuji process	400	HZSM5	PE, PP, PS, PET, PC	85% oil

Appendix D

See Table 8.

Table 8 Industrial facilities for plastic hydrocracking. *Source:* Adapted from Samperio (2016).

Process	Temperature, °C	Feedstock	Yield
Hiedrierwerke Freiberg Böhlen ITC	400 400–435 450–470 435	PW PW PW	oil and gas oil, gas and solid residue 80% oil oil and gas

Appendix E

See Table 9.

Table 9Industrial facilities for plastic gasification process. *Source:* Adapted from Nistelrooij (2017) and Samperio (2016).

Process	Temperature, °C	Feedstock, %	Yield
Ebara, TwinRec	500-600	PW, ARS	energy and metals
Texaco	1200-1500	PW (10)	gas
Lurgi (SVZ)	1600-1800	PW, ARS, waste oil, lignite, WEEE	gas, CH₃OH, energy
Enerkem	unknown	household waste	oil, alternative fuels

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