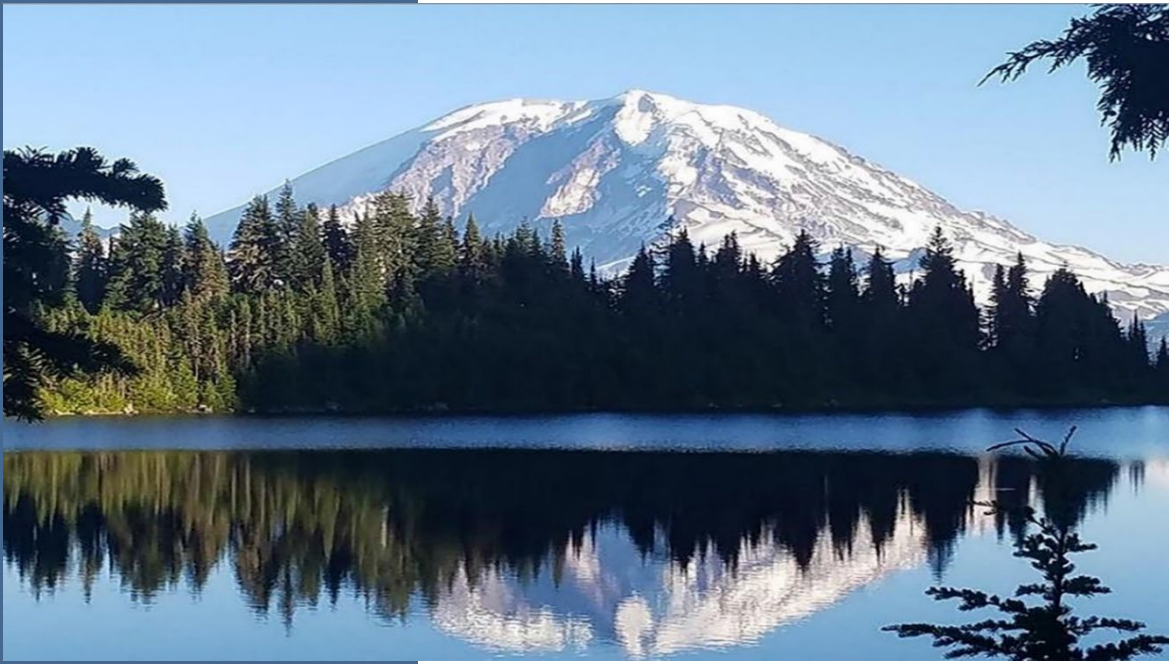

Evaluation of Emerging Plastics

Recycling Technologies and Management Strategies

By

Sohrab Haghighi Mood, Georgine Yorgey, Manuel Garcia-Perez

Solid Waste Management Program

Washington State Department of Ecology

Olympia, Washington

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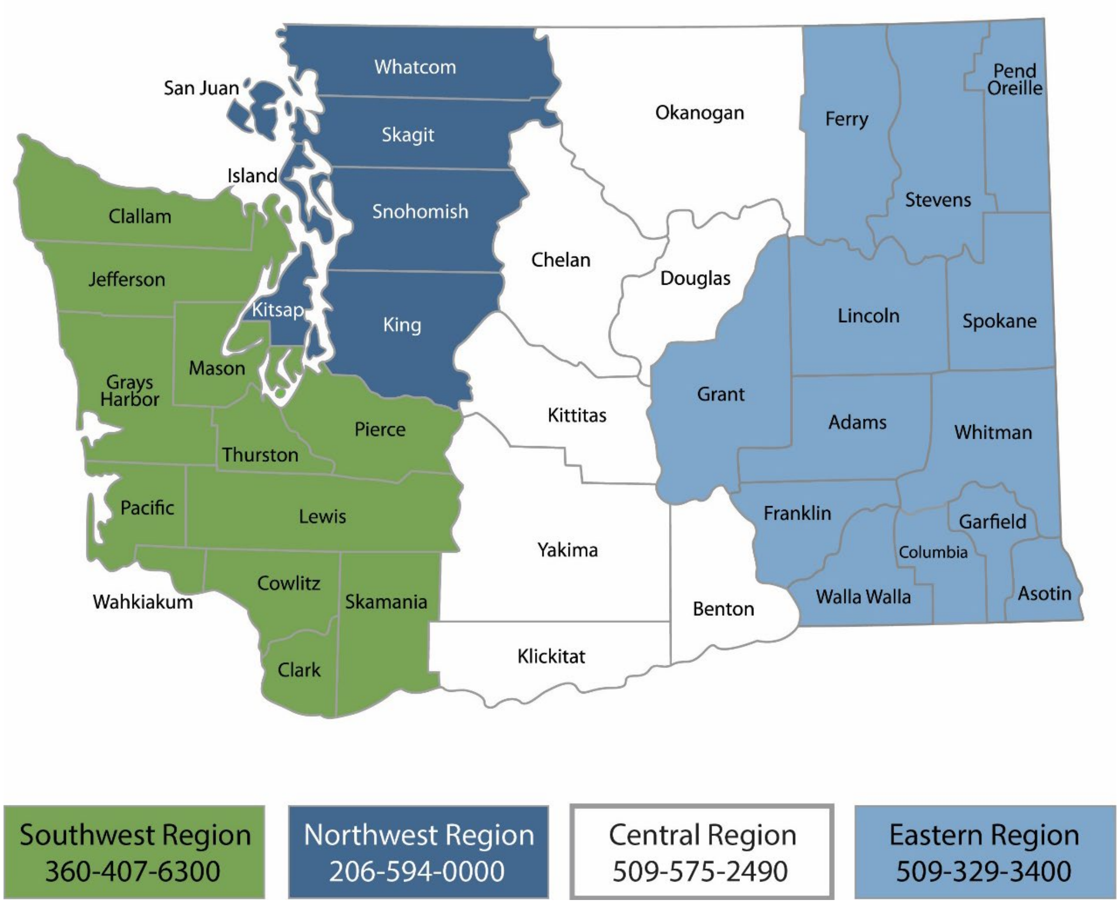

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DEPARTMENT OF
ECOLOGY
State of Washington

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Abstract

Plastic waste is a growing concern both within Washington State and globally because of high production, high durability, and the lack of suitable waste management systems. Most plastic waste is currently landfilled, and when plastic waste is released into the environment, it can break down into micro-plastics that can have negative impacts on ecosystems. Mechanical recycling could be a solution, but practically speaking, because of the relatively high purity requirements for new plastics production, this method recycles only a fraction of plastic waste. Mechanical recycling also generally downscales the material, with recycled materials having lower performance than the original ones. Therefore, there has been interest in chemical recycling as an additional recycling option. This literature review identifies the most significant, appropriate, and scalable technologies currently being promoted for chemical recycling applications, including gasification of municipal solid waste (MSW), pyrolysis of MSW, and carbonization of MSW. Many of these technologies are mature and are being newly applied to plastic feedstock. We explain each approach's major strengths and weaknesses to the extent this information is available in the literature. We also describe the plastics conversion products of each technology (fuel, upcycled plastics, downcycled plastics, etc.) and how these products might be integrated into more holistic waste management approaches that support a more circular economy. Given the burgeoning interest and research into bio-based plastics, these recycling technologies' applicability for bioplastics are also reviewed. Two parallel questions are therefore being considered: how to handle best the plastic waste that is currently creating management issues for solid waste personnel, and how these issues could change in the future given different policies, technologies, plastic types, and waste management approaches.

1. Introduction

Plastics are widely used in many applications due to their low manufacturing cost and high durability, strength, mouldability, and versatility (van Emmerik and Schwarz, 2020). However, although plastics combine unrivaled functional properties and low costs that have substantially contributed to living standards, the need for appropriate disposal negatively impacts the environment (Lopez, Artetxe et al. 2017). During disposal and landfilling, harmful materials in plastic waste can be released into the soil and groundwater, leading to environmental concerns (Mahadevan Vaishnavi and Kannappan Panchamoorthy Gopinath 2023). Plastic waste has strong biological and chemical stability, and it does not decompose easily (Xiao, Yu et al. 2023). As plastics in rivers and marine environments are exposed to wind, rain, and sunlight, the structure becomes more susceptible to fragmentation into microplastics(Chen, Liu et al. 2020). Microplastic can adsorb organic pollutants, heightening toxicity (Chen, Liu et al. 2020). With nearly a third of all plastics leaking products into the environment, by 2050, there could be more plastics than fish in the oceans (Lopez, Artetxe et al. 2017).

Plastic waste has grown substantially over the last sixty years; in 2018, 35.7 million tons of plastics were generated in the United States, accounting for 12.2 percent of MSW generation (Figure 1) (United States EPA 2022). Of this, about 9% is recycled, and an additional 16% is incinerated with energy recovery.

In 2017, 410,300 tons of plastic packaging waste were generated by residents and businesses in the state of Washington, 112 pounds of plastic packaging waste per person per year (Ecology 2017, Ecology 2020). Approximately 17 percent of the total plastic packaging generated (69,410 tons) was reprocessed by recycling in the state of Washington(Ecology 2020).

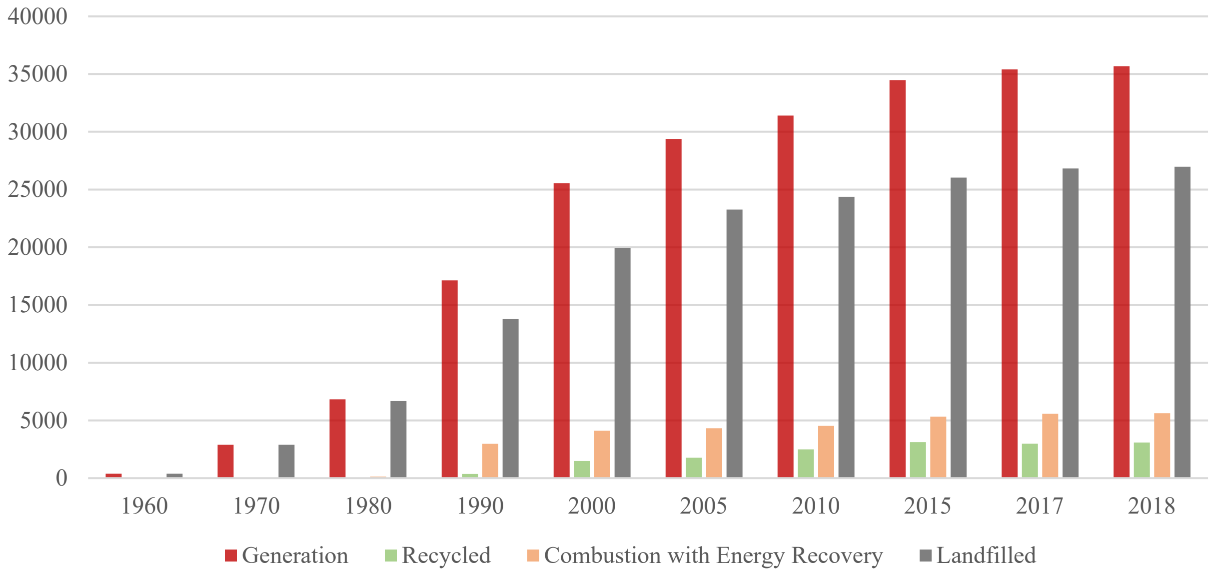

Figure 1. 1960-2018 Data on Plastics in MSW by Weight (in thousands of U.S. tons) (United States EPA 2022)

To date, humans have produced more than 8,300 million metric tons (MT) of virgin plastics, mostly from fossil hydrocarbons (Geyer, Jambeck et al. 2017). Of this quantity, 6,300 MT have been disposed of, while between 2,000 and 2,500 million metric tons (MT) are still in use forming part of our infrastructure. Plastic packages are of special concern; after a single use, more than 95% (with an estimated value of $ 80-120 billion annually) go to landfills (Lopez, Artetxe et al. 2017). Figure 2 shows the current supply chain to produce virgin plastics; natural gas and naphtha are the two major feedstocks (Li, Aguirre-Villegas et al. 2022).

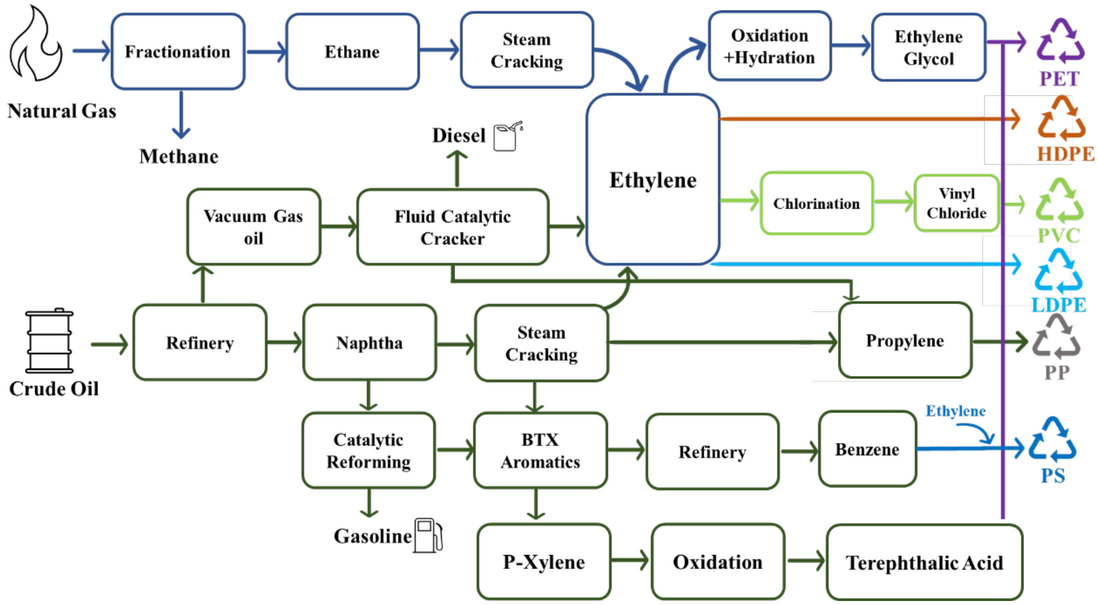

Figure 2. The supply chain for the manufacture of virgin plastics ((Adapted from (Li, Aguirre-Villegas et al. 2022))

The transformation of a linear product production and disposal system to a more circular one can reduce the waste stream by converting portions into energy or valuable products (Figure 3)(Vaishnavi, Vasanth et al. 2023). Figure 4 shows a simplified scheme depicting current plastics management and disposal challenges and how this new paradigm could address some of these challenges. The new paradigm targets the conversion of hard-to-recycle plastic to high-value materials and fuels with minimum landfill discharge.

There are many reasons why many plastics are landfilled rather than recycled. Landfilling represents the least expensive and easiest method for management (Li, Aguirre-Villegas et al. 2022). In addition, recycling capacity is not able to handle the amount and types of plastic we are disposing of. Post-consumer plastics are intrinsically heterogeneous and made of different polymers (e.g., mainly PE, PP, and PET) as well as foreign materials such as foreign polymers, additives(e.g., dyes and fillers), and contaminants (Lange 2021).

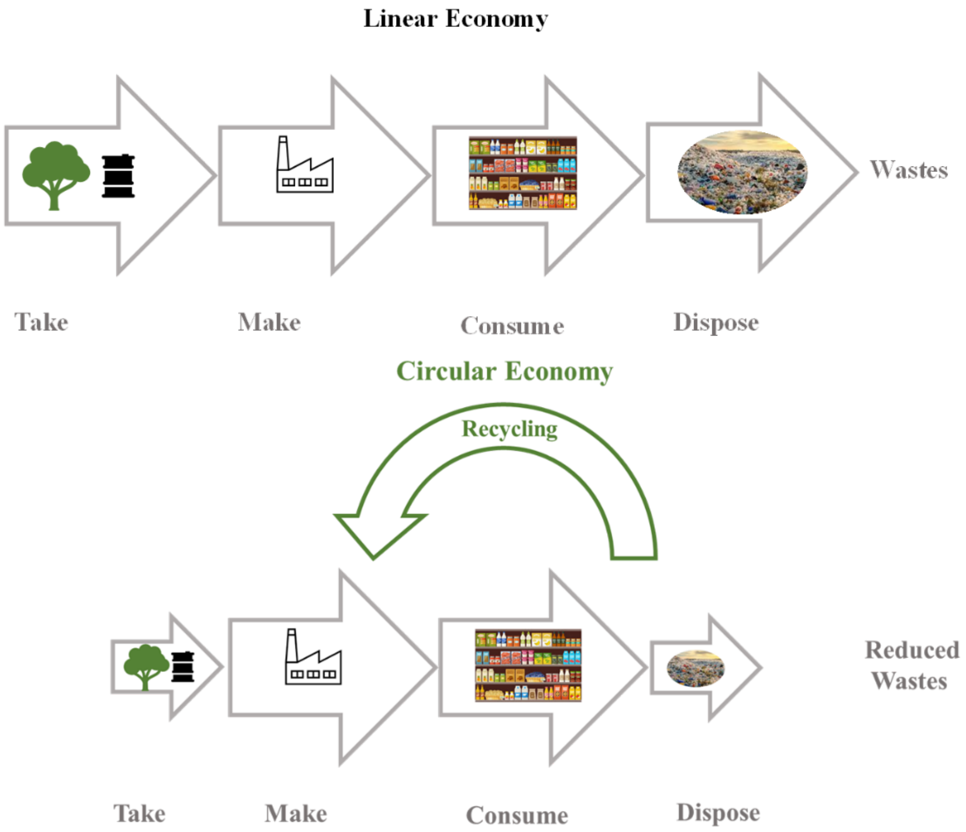

Figure 3. Comparison of the circular and linear economy showing the product life cycle

Figure 4. Current situation with plastics utilization and our targeted paradigm (Adapted from Geyer et al., 2017).

The 3 Rs concept (Reduce, Reuse, Recycle) can be built upon to help transform the linear plastics paradigm into a more circular one. "Regeneration" (sometimes also called “Repurposing”) represents a fourth R that addresses material transformation, providing added value to waste material and exploring high-value production materials, which enhance economic benefits (Roy, Garnier et al. 2021) (Figure 5).

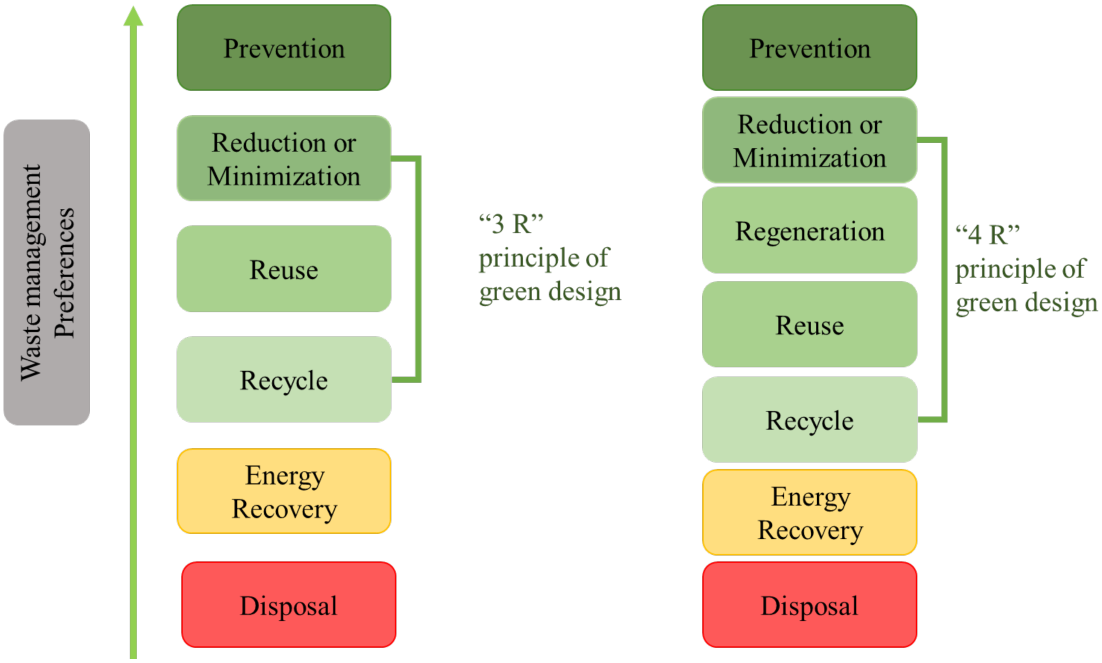

Figure 5. Waste management hierarchy incorporating the "3R" and "4R" concepts. (Adapted from Roy et al., 2021).

Among the 4Rs, Recycling is one of the main strategies traditionally used for achieving a circular economy (Islam, Iyer-Raniga et al. 2022), though it is important to note that it is at the midpoint of the hierarchy of plastics management (Figure 5). Traditional recycling, primarily revolving around mechanical techniques, is hampered by many (and sometimes complex) processing steps, which include collection, identification, sorting, grinding, washing, separating, volume reducing, extruding / compounding, and pelletization (Chen, Liu et al. 2020). Recycled plastics have lower economic value compared to original plastics due to the presence of contamination and impurities in plastic wastes (Chen, Liu et al. 2020); thus, they can be considered "downcycled." Advanced or Chemical recycling received much attention recently for its potential to efficiently convert more of our plastic waste(Englund, Li et al. 2021).

Though lower on a waste hierarchy still, incineration (with energy recovery) could be a suitable method to take advantage of the high calorific value of plastics; however, the emission of harmful gasses such as dioxins and furans needs to be accounted for and limits its use (Katami, Yasuhara et al. 2002). Greenhouse gas (GHG) emissions from the incineration of plastic waste is estimated to be 1.8–3 kg $CO_{2,\,eq}$ per kg plastic waste (Rudolph, Kiesel et al. , Khoo 2019, Li, Aguirre-Villegas et al. 2022). Moreover, the solids that remain after incineration contain a high amount of microplastics, which is harmful to the environment (Jiang, Shi et al. 2022).

Within this context, chemical and advanced recycling technologies (Gilbert 2016, Tullo 2020) include a broad range of technologies that include chemical, thermal, or biological process types. Applying chemical, thermal, and biological technologies can convert plastics waste into different potential products such as fuels, lubricants, monomers, new polymers, and carbonaceous materials (Lopez, Artetxe et al. 2017, Martín, Mondelli et al. 2021, Hinton, Talley et al. 2022). The process of converting plastic to fuel is often considered energy recovery rather than recycling. However, if the final product is plastic which will subsequently be used as fuel, the process is considered plastics-to-plastics, or material-to-material, and is typically seen as a type of recycling (Product Stewardship Institute 2022). Despite these limitations, with further technology development, chemical and advanced recycling technologies are promising technologies for the future due to their ability to produce valuable chemicals and fuels from plastic waste, which is consistent with the 4R concept.

1.2 Waste Sorting

The first steps in plastic recycling are collection and separation. Plastic wastes are generally sorted through sequential steps (Ragaert, Delva et al. 2017, Serranti and Bonifazi 2019, Lange 2021), which include sorting by size, eliminating foreign materials such as metal and glass, sorting by type of plastic material, sizing, and finally, granulation into plastic recyclate (Lange 2021). Depending on the composition of the wastes, foreign materials can be separated by various methods, such as air classifiers using gravity or sink float (Figures 6A and B). Metals can be separated using the magnetic attraction of ferrous metal or through induced magnetic repulsion of nonferrous metals (Lange 2021)(Figure 6 C). An infrared (IR) detector is also often used to identify and separate different plastic wastes (Figure 6 D). The standard IR detector can be substituted or complemented by hyperspectral imaging spectroscopy (HIS) to identify a full-shape product or by an X-ray fluorescence detector to identify heavy elements such as Cl and Br (Lange 2021). Recently, hyperspectral imaging (HSI), which combines imaging from a digital camera with spectrometric analysis, has been used to get a discrete spectrum for every pixel collected, which can be analyzed computationally for the sorting of more complex plastic streams (Li, Aguirre-Villegas et al. 2022). Under current sorting technologies, manual sorting is usually used after these techniques to address sensor errors and other drawbacks of these techniques (Ragaert, Delva et al. 2017).

This progress in sorting and collecting plastic waste could improve options for mechanical and chemical recycling and energy recovery from plastic waste. New technologies are also being developed that may further ease sorting, such as tracer-based sorting using fluorescent pigments incorporated into the plastic substrate and technology to apply digital watermarks (e.g., codes) that could be integrated into the packaging design and which can then be identified by cameras on high-speed sorting line (Lange 2021). Sorting is an important step in plastic waste management. However, it is not usually sufficient for recycling, as it does not address plastics that may contain dirt and other contaminants.

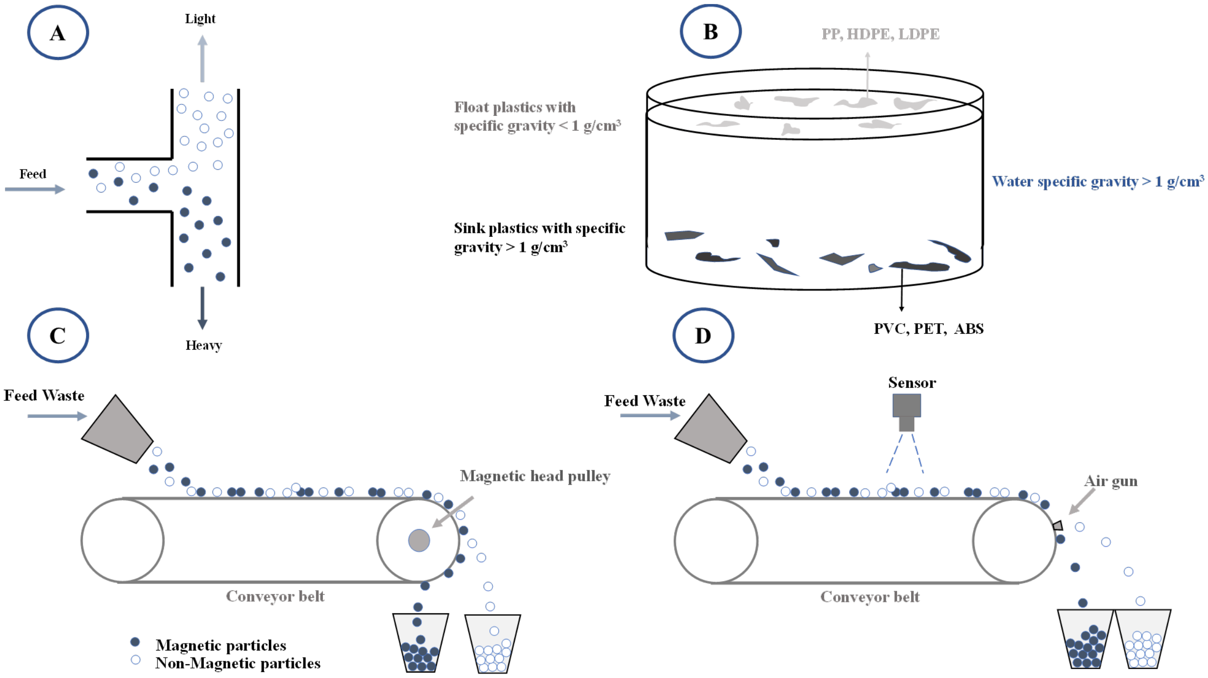

Figure 6. Sorting technologies: A, air classifier; B, sink-float sorting; C, magnetic sorting of ferrous metals; D, sensor-based sorting (Adapted from Serranti and Bonifazi 2019).

2. Mechanical Recycling

The most common recycling method for plastics is mechanical recycling. Thus, though it is not the focus of this review, it is worth understanding as an important point of comparison. In an important sense, it is the limitations of mechanical recycling which have driven the interest and further development of chemical recycling. The scheme of the main steps in a mechanical recycling process is shown in Figure 7.

In mechanical recycling, plastics are melted or softened; however, the polymers with long chain molecules are not broken down and they are reformed into a pellet, aggregate, or its final shape(Englund, Li et al. 2021).

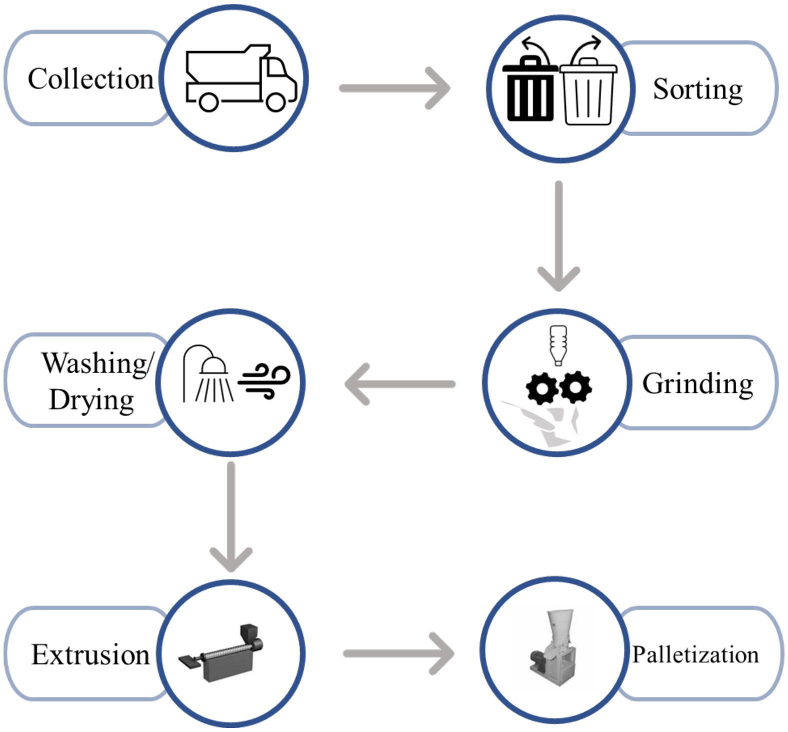

Figure 7. The scheme of the main steps in a mechanical recycling process

Mechanical recycling cannot be used for all types of plastics. For instance, thermoset plastics are among those plastics that cannot be mechanically recycled since their chemical structure prevents melting (Davidson, Furlong et al. 2021).

Mechanical recycling has focused mainly on the three dominant packaging polymers PE, PP, and PET (Lange 2021). A diverse plastic waste stream generally cannot be mechanically recycled as a single stream due to thermodynamic incompatibilities and mixed properties (Maris, Bourdon et al. 2018, Hinton, Talley et al. 2022). The differences in melting points and processing temperatures between the polymers in mixed plastics streams make recycling challenging. The mechanical recycler usually sets the processing temperature of the highest melting component, which causes overheating and degradation of some lower melting components, and leads to lower properties of the material (Ragaert, Delva et al. 2017). For example, high processing temperatures applied for PET result in degradation and dehydrochlorination of the PVC (Möller and Jeske 1995).

Mechanical recycling processes thus depend on careful sorting of the clean and pure plastics into mono-stream fractions. These fractions are then compounded into granules, and mixed with a virgin polymer of the same family, together with compatibilizers and additives to mitigate the shortcomings of recycled material (Ignatyev, Thielemans et al. 2014, Ragaert, Delva et al. 2017, Lange 2021). Currently, mechanical recycling is usually a form of downcycling, with roughly 10% material quality loss in each cycle of processing. The changes in plastic structure can be challenging for mechanical recycling (Ragaert, Delva et al. 2017). Plastics can only be recycled mechanically about seven times before the polymers are too degraded for further use (Product Stewardship Institute 2022).

Loss of quality during mechanical recycling is a result of several factors. First, plastics are exposed to heat, oxygen, light, radiation, moisture, and mechanical stress during their lifetime, which causes a photo-oxidation process in plastics (Ragaert, Delva et al. 2017), resulting in the formation of oxygenated groups on the polymer chain. Second, during mechanical recycling, plastics are degraded due to heat and mechanical shear used (Ragaert, Delva et al. 2017). Third, sorted plastic wastes don’t have the purity of virgin materials, and always traces of different polymers can be observed in sorted plastic which lowers the quality of recycled materials. Contaminants that are not completely soluble can induce phase separation, which negatively impacts mechanical properties(Al-Salem, Lettieri et al. 2009, Ragaert, Delva et al. 2017). And fourth, when compatibilizers and additives are blended with recyclate and virgin resin to overcome some of these shortcomings, the level of impurities in the recycled resins increases (Lange 2021).

Mechanical recycling is not a suitable option for some applications. For example, mechanical recycling of thermoset materials such as polyurethane mattresses and vulcanized rubber tires is very difficult. Meanwhile, mechanically recycled products cannot be used to make materials for food applications such as food packaging if they might be contaminated by traces of toxic impurities (Lange 2021).

The plastic products' design greatly impacts both their recyclability and the degree to which they can incorporate recycled materials(Ragaert, Delva et al. 2017). Products should be made in ways that they can be recycled easily. The recycled materials should be fully characterized to identify the recycled polymer’s strengths and weaknesses (Ragaert 2016).

2.1 Key Messages for Mechanical Recycling

- The most common recycling for plastics is mechanical recycling.
- Mechanical recycling is not a suitable method for mixed plastic waste. Rather, it relies on plastics that have been successfully separated by polymer type.
- Mechanical recycling of plastics causes material quality loss in each cycle of processing.

3. Chemical/Advanced recycling processes

In the United States, around 40 companies are working on developing chemical recycling projects (Table A1). More chemical recycling plants are being established, with the idea that these processes can complement or even ultimately replace mechanical plastic recycling if chemical recycling methods are developed to have environmentally friendly processes (in terms of both waste and emissions) and produce higher-value products. To achieve this, the products of chemical recycling technologies will need to avoid the generation of harmful pollutants and have lower lifecycle effects compared to traditional means (Jeswani, Krüger et al. 2021).

Chemical recycling is also of interest because of the products that it can generate. Demand for post-consumer recycled resins has increased over the past years, mainly due to policies enacting post-consumer recycled content requirements for certain types of plastics(e.g., food-grade and bottle-grade packaging) (Product Stewardship Institute 2022). Many industry stakeholders believe that chemical recycling is the only feasible method for achieving both post-consumer recycled content requirements and state and federal health and safety requirements for food-grade applications (Product Stewardship Institute 2022).

Chemical recycling processes include a range of processes that apply one of three technology types: purification/additive extraction, depolymerization, or thermal conversion. Though these processes are used at commercial scales, it is still not entirely clear how to classify these processes. For example, The American Chemistry Council and other industry groups consider purification/additive extraction, depolymerization, and thermal conversion technologies as manufacturing processes since they use waste plastics as feedstocks to produce either fuels or the building blocks for new plastics(Product Stewardship Institute 2022). However, environmental groups support regulating these technologies as waste management processes since waste management facilities have more stringent restrictions on emissions (Product Stewardship Institute 2022).

3.1 Purification /Additive Extraction

In purification/additive extraction (which we will refer to as purification), plastics are dissolved in chemical solvents to remove additives and dyes from plastic waste and recover virgin-grade plastic resins (Ügdüler, Van Geem et al. 2020, Vollmer, Jenks et al. 2020, Product Stewardship Institute 2022). The selection of solvents is an important factor in the feasibility of the process. The waste polymer is washed with solvents or supercritical fluids – where supercritical fluids are substances at a temperature and pressure above their critical point, where distinct liquid and gas phases do not exist but below the pressure required to compress it into a solid (Ügdüler, Van Geem et al. 2020). Compared to other chemical recycling technologies, purification is the least energy-intensive and has the highest plastic-to-plastic processing efficiency rate (Product Stewardship Institute 2022). On the downside, the recovery of solvent and antisolvent can be energy-intensive and costly (Lange 2021). Purification can be employed for single-material plastics (also called mono-material plastics) such as polypropylene, polyethylene, polyethylene terephthalate, and polystyrene. These processes work well with source-separated and clean plastic waste. The product of purification is virgin-like plastics of the same polymer type as feedstock (Product Stewardship Institute 2022). Solvent recovery is an important factor in the economics of the additive extraction process, and recovery rates can be achieved up to 100% (Ügdüler, Van Geem et al. 2020). This method may also need large amounts of solvent, which causes challenges related to toxicity and energy consumption (Jessop 2011, Ashcroft, Dunn et al. 2015, Li, Aguirre-Villegas et al. 2022).

PureCycle Tech is one example of a company that uses a plastics-to-plastics purification technology to separate color, odor, and other additives and contaminants from Polypropylene to "transform it into a virgin-like resin"(Product Stewardship Institute 2022, Purecycle 2023). The scheme of the main steps occurring in Purecycle technology processes is shown in Figure 8.

Figure 8. The scheme of the main steps occurring in Purecycle technology processes

3.1.1 Summary of Key Messages

- The purification method can recover virgin-grade plastic resins.
- The recovery of solvents in the purification process can be energy-intensive and costly.

3.2 Depolymerization

Depolymerization is a reverse polymerization reaction. Molecular bonds of plastics are broken to recover building blocks of monomers or oligomers that can be repolymerized into resins (Figure 9) (Davidson, Furlong et al. 2021). Biological, chemical, or thermal means or a combination of these methods can be applied to break molecular bonds (Webb, Arnott et al. 2013). Depolymerization is reported to be more energy intensive than purification and less energy intensive than thermal conversion (Product Stewardship Institute 2022). Compared to purification, depolymerization can process a wider variety of materials with higher levels of additives and contaminants. Depolymerization also needs a degree of pre-processing (e.g., clean, mono-material inputs).

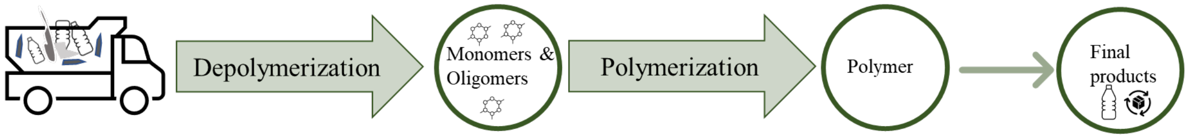

Figure 9. A scheme representing chemical depolymerization processes.

3.2.1 Chemical depolymerization

In this method, chemical reagents are employed to decompose plastics into their building blocks. Some examples of chemical depolymerization technologies include hydrolysis, methanolysis, and glycolysis, which are depolymerizing plastics in a water-based solution, in methanol, and in glycol, respectively (Alberti, Damps et al. 2019, Ügdüler, Van Geem et al. 2020). Specific mono-material polymers (e.g., condensation polymers) are used for chemical depolymerization. This process can accept some contamination with additives, pigments or colorants, and non-target polymers. Monomers or oligomers generated in this process can be used to produce new plastic. Reaction parameters (e.g., reaction time, temperature, catalyst) can affect efficient depolymerization (George and Kurian 2014). Sub- and supercritical fluids such as water and alcohol can be used to depolymerize or decompose plastics to monomers without using catalysis (Ragaert, Delva et al. 2017).

Several existing companies use depolymerization technologies to recycle plastics. Matsushita Electric Works, located in Japan, applies a technology using hydrolysis in subcritical water for the depolymerization of flame-retardant polymers (Ragaert, Delva et al. 2017). The thermosetting resin in flame-retardant polymers can be recycled into basic materials with a material recycling rate of 70% (Ragaert, Delva et al. 2017). Eastman Company, located in Tennessee, has developed polyester renewal technologies that apply chemical depolymerization using glycolysis and methanolysis to produce monomers of polyester, with a primary focus on methanolysis. The monomers produced in this process can be used to generate co-polyesters, plastics, and chemicals. This technology can reduce greenhouse gas (GHG) emissions by 20-30% as compared to fossil-fuel-based production of the same monomers (Product Stewardship Institute 2022).

3.2.1.1 Summary of Key Messages

- Chemical reagents are employed to decompose plastics into their building blocks which can be further processed into new plastics and chemicals.
- This process can accept some contaminations.

3.2.2 Thermal depolymerization

This method decomposes plastics into monomers or oligomers by heating. Supercritical conditions, or the use of catalysts, are usually needed to improve the efficiency of the depolymerization process (Jehanno, Pérez-Madrigal et al. 2019). Monomers and oligomers can be subsequently repolymerized into polymers, or monomers can be applied as high-added-value building blocks for producing new materials or chemicals (Jehanno, Pérez-Madrigal et al. 2019). Thermal depolymerization can be combined with chemical processes. Thermal depolymerization is applied to polymers such as Polypropylene, Polystyrene, and acrylics. The operating conditions need to be carefully controlled to maximize process efficiency and minimize unwanted degradation of products (Newborough, Highgate et al. 2002).

Agilyx, located in Tigard, Oregon, treats post-consumer and post-industrial mixed plastics using different technologies, including thermal depolymerization (Product Stewardship Institute 2022).

3.2.2.1 Summary of Key Messages

- Heating is employed to decompose plastics into their building blocks which can be further processed into new plastics and chemicals.
- The operating conditions need to be carefully controlled to maximize process efficiency and minimize unwanted degradation of products.

3.2.3 Biological depolymerization

In this method, enzymes and microorganisms are used instead of chemical solvents or heat to deconstruct plastics into their monomers or oligomers. Biological depolymerization is a slow process(Ali, Bukhari et al. 2023). Few biological depolymerization technologies(enzymatic) are available, and those that do exist are primarily used for processing PET, from textiles and beverage bottles. Natural hydrolase enzymes have been used successfully for polymers with structures that are similar to the natural macromolecules, such as poly(hydroxy alkanoates) (PHAs)(Hinton, Talley et al. 2022).

Carbios, located in France, has developed an enzymatic recycling technology for PET. They use enzymatic hydrolysis to decompose PET from rigid plastics, along with textiles, into the monomers PTA and EG (ethylene glycol) (Carbios 2022). Bioxyle, located in Atalanta, US, develops enzymes and processes that enhance the biodegradation of plastics back to their original state (Bioxycle 2023).

3.2.3.1 Summary of Key Messages

- Enzymes and microorganisms are used to deconstruct plastics into their monomers or oligomers.
- Biological depolymerization is a slow process.

3.2.4 Overall Summary of Key Messages for Chemical Depolymerization

- Compared to mechanical recycling, recycled plastic obtained from the depolymerization process can have virgin-like plastic properties.
- Depolymerization processes can produce a broader range of materials which include those with higher levels of additives and contaminants compared to the mechanical recycling method.

3.3 Thermal Conversion

Technologies such as pyrolysis and gasification are alternative methods for processing plastic waste and can be used to produce fuel or fuel intermediaries, which can be further processed into refined hydrocarbons, new plastics, or other petrochemicals. The main difference between the products of depolymerization and conversion is that liquid, gaseous hydrocarbons, and char are the products of thermal conversion technologies, whereas plastic monomers or oligomers are the products of depolymerization. The main advantage of thermal conversion technology is that a wider range of plastics, such as contaminated mixed materials and durable, bulky plastics, can be used as feedstock.

3.3.1 Pyrolysis

Pyrolysis is used to convert organic materials and plastics into energy and chemicals in an oxygen-free environment at high temperatures (e.g., 300-700° C) (Haghighi Mood, Pelaez-Samaniego et al. 2022). The products of pyrolysis of plastics include oils, waxes, gases, and char. A variety of hydrocarbons can be found in pyrolysis oil, including olefins, paraffin, aromatics, diolefins, iso-paraffins, and naphthenes (Li, Aguirre-Villegas et al. 2022). The product distributions are dependent on feedstock, reaction conditions, and reactor type(Li, Aguirre-Villegas et al. 2022). Plastic pyrolysis can convert plastics into lower molecular weight products, which can be used as fuels or feedstock for producing chemicals or other plastics. Pyrolysis generates less emissions compared to incineration (Jeswani, Krüger et al. 2021). In contrast to mechanical recycling, pyrolysis can handle heterogeneous plastic mixtures and highly contaminated feedstocks such as automotive shredder residue (Vermeulen, Van Caneghem et al. 2011). This technology could therefore be an appropriate method to manage plastic waste that is challenging to depolymerize, and which cannot be mechanically recycled – where the only alternative strategies are to incinerate or landfill them. Examples of this type of waste stream include mixed PE/PP/PS, multi-layer packaging, fiber-reinforced composites, polyurethane construction and waste from demolition during construction (Ragaert, Delva et al. 2017). A typical mixed plastics sample from a material recovery facility may also contain a significant fraction of contaminants, including latex, medical waste, paper, clothing waste, wood waste, glass, inks, pigments, and metals (Hopewell, Dvorak et al. 2009, Adrados, de Marco et al. 2012).

Regarding air pollution, pyrolysis can provide improvements over incineration as the emission of organic compounds in the incineration of plastic waste is ten time higher than that of the air emission of pyrolysis (Li, Aguirre-Villegas et al. 2022).

The products of plastic pyrolysis include oils and waxes, gases, and char. The oils and waxes can be either burned as fuel or converted to plastic monomers, followed by repolymerization or other processes to produce polymers and fuels. The energy content of oils obtained via pyrolysis can range from 41.10 to 46.16 MJ kg^{-1}, which is in the range of the heating values of conventional fuels (Antelava, Jablonska et al. 2021). Oil composition varies based on feedstocks, but additional treatment is usually needed to maintain the oil quality. For example, sulfur should be separated from the oil to meet market standards if it is present (Antelava, Jablonska et al. 2021). Meanwhile, the gas generated during the pyrolysis process can be used to generate electricity. Pyrolysis can connect the circular economy value chain by connecting waste management operations with new manufacturing operations. Upstream pyrolysis, waste collection, aggregation, and sorting operations take place. Downstream, other entities utilize the products of pyrolysis as fuel or to produce new plastics or chemicals. An example of a value chain is shown in Figure 10.

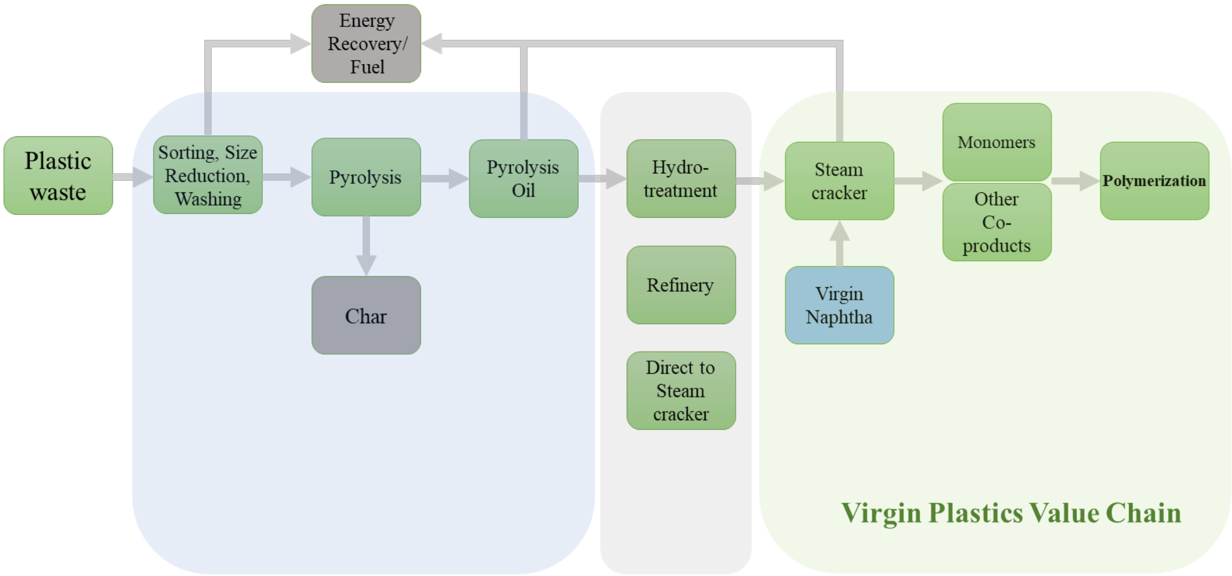

Figure 10. Pyrolysis within the Advanced Recycling Value Chain (Adapted from Gendell et al., 2022)

Waste plastics vary from one another and differ from model plastics regarding structures, constitutions, properties, etc. Pyrolysis of LDPE and HDPE resulted in about 43% of liquid oil, with the remaining products of gas and waxes (Miskolczi, Wu et al. 2016). Pyrolysis oils from PE and PP are rich in aliphatic hydrocarbons; however, aromatic compounds are much easier to convert into carbons. Even pyrolysis has feedstock specifications that need to be met. For example, even pyrolysis facilities do not cope well with film plastics (e.g. bags and wraps), which can wrap around equipment and cause issues during sorting and plastics movement. These specifications are described more fully in Appendix B. The economic performance of the pyrolysis technology is necessary for developing pyrolysis products refineries as investors expect profitable projects. Li et al provided the summary of waste plastic techno-economic analysis pyrolysis articles by feedstock, products, region, capacity, capital cost, and return on investment in appendix B (Li, Aguirre-Villegas et al. 2022).

Co-pyrolysis of biomass with plastics or other hydrogen-rich feedstock has received much attention since adding plastic to biomass for pyrolysis can enhance the quality of the bio-oil by increasing the yield of aromatics and reducing the coke formation (Zulkafli, Hassan et al. 2023). This strategy could help cope with the lower effective carbon-to-hydrogen ratio in pyrolysis of biomass alone responsible for the formation of oxygenated compounds, which can be eliminated by dehydration, decarboxylation, and decarbonylation reactions and aid in enhancing the bio-oil quality (Alam, Bhavanam et al. 2020, Dada, Islam et al. 2021, Mahadevan Vaishnavi and Kannappan Panchamoorthy Gopinath 2023).

Nexus Circular company**,** located in Atlanta, Georgia, uses pyrolysis to convert post-industrial and post-commercial plastics to oils and waxes, which are further processed to produce like-new polyethylene resin. This resin can be further converted into new plastic (Nexus Circular).

Agilyx, located in Oregon, uses pyrolysis technology and purification techniques to create a variety of products from hard-to-recycle plastics(Agilyx).

Anellotech, located in Pearl River, New York, developed a catalytic process called “Plas-TCat” to convert mixed plastic waste into olefins and aromatics, and the produced products are applied as “drop-in” raw materials to produce new plastics and work towards a plastic circular economy(Li, Aguirre-Villegas et al. 2022).

3.3.1.2 Summary of Key Messages

- Pyrolysis operators can use a mix of plastics and colors and have a different set of considerations about contaminant threshold limits than mechanical recyclers. However, high amounts of contaminants can lead to undesirable effects such as lowered process yield, reduced output quality, and wear on equipment.
- Pyrolysis of plastics has less emission of organic compounds compared to incineration.
- A variety of gas, liquid, and gas products can be produced using pyrolysis technology.
- Landfills need to evaluate different sorting strategies to obtain feedstocks with predictable properties amenable to standardization.
- The impact of impurities in MSW fractions on targeted conversion technologies should be thoroughly studied.

3.3.2 Gasification

Gasification processes materials thermally under a controlled oxidizing environment at high temperatures (above 700 °C). Gasification has better flexibility regarding feedstock composition than pyrolysis (Lopez, Artetxe et al. 2018). Figure 11 shows a general flow diagram for waste plastic gasification processes(Li, Aguirre-Villegas et al. 2022). As the gasification process begins, the volatiles are exposed to physical, thermal, or chemical gas cleaning technologies in which char, slags, and ash are separated. The main product of gasification of waste plastics is syngas comprised of H_2, CO, CO_2, CH_4, and N_2. The clean syngas produced during gasification (Figure 11) is cooled using a quench followed by secondary gas cleaning step. The secondary gas cleaning separates contaminants that can affect catalyst performance (Gogate 2019, Li, Aguirre-Villegas et al. 2022). Syngas can be used for producing energy, energy carriers (e.g., H_2), and chemicals (Saebea, Ruengrit et al. 2020). Methanol and transportation fuels can be produced from syngas. Methanol can subsequently be converted to aromatics and olefins (Ilias and Bhan 2013). Aromatics such as benzene, toluene, and xylenes mixture are extensively used as raw materials to produce resin, rubber, and fiber(Fu, Guo et al. 2022). Olefines can be used to produce many plastic materials. Biooil and biochar are other products of the gasification process (Patra, Patra et al. 2022).

Several factors, including feedstock concentration, oxidizing agent, temperature, and time, affect the gasification process, leading to different qualities and quantities of the produced syngas (Antelava, Jablonska et al. 2021). Feedstock composition and blend of feedstock with biomass could affect the products. For example, the blend of plastics with woody biomass showed the production of more H_2 and CO, and less methane and other light hydrocarbons compared to the gasification of plastic(Zaccariello and Mastellone 2015). Moreover, oxidizing agents can affect the Producer gas. For example, CO, H_2 and N_2 are the main gases produced when air is used for gasification. However, CO and H_2 are major gases produced when oxygen is used for gasification(Li, Aguirre-Villegas et al. 2022). More information about the gasification process is provided in Appendix C.

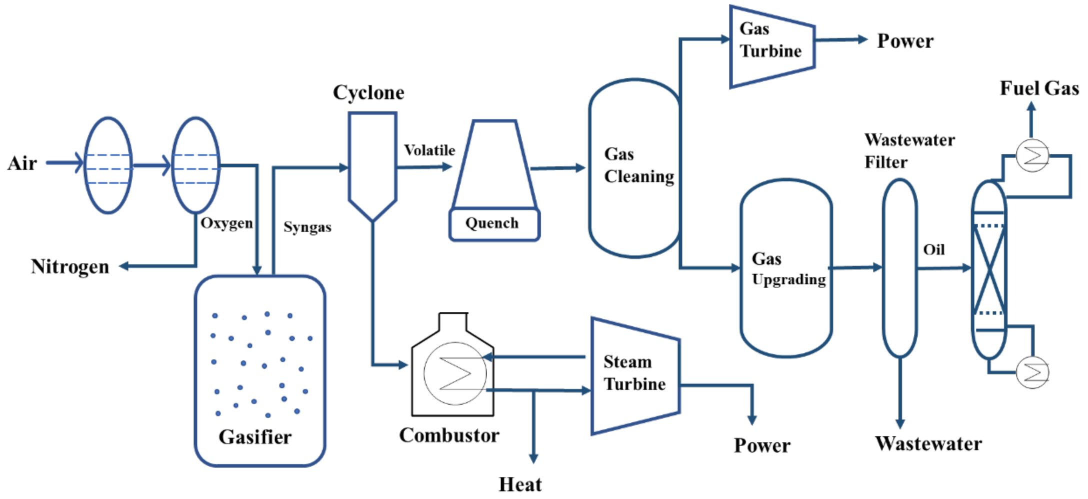

Figure 11. Gasification of waste plastics and upgrading to heat, power, olefins, alcohols, and other fuels (Adapted from Li et al., 2022).

Gasification technology is suggested to be more profitable and environmentally sustainable compared to incineration(Arena, Mastellone et al. 2003, Nelson 2007). A study shows that the global warming effects of waste gasification with a combined cycle powerplant are over 50% less than incineration(Dong, Tang et al. 2018, Li, Aguirre-Villegas et al. 2022).

Eastman Company, located in Tennessee, uses different types of plastic waste as feedstock to produce syngas, which is used to replace coal-based syngas feedstocks for plastics, paint additives, and textile fibers., Eastman's carbon renewal technology could decrease the GHG emissions for the production of syngas by 20% to 50% (Eastman 2020).Shell and its collaborators used Enerkem's gasification technology to convert MSW to syngas and further process to produce methanol(Enerkem 2021).

3.3.2.1 Summary of Key Messages

- Gasification has better flexibility regarding feedstock composition than pyrolysis.
- A wide range of products can be obtained using gasification technology.
- Global warming effects of waste gasification are much less than incineration.

3.3.3 Carbonization

The carbonization of plastic wastes is the conversion of plastics to carbon materials, such as carbon fiber and carbon nanotubes, conducted through a high-temperature heat-treatment process under different operational conditions (e.g., oxidative or inert atmosphere, atmospheric or high pressure)(Chen, Liu et al. 2020).

Plastic carbonization can be generally divided into two major steps: (1) thermal degradation (in the presence of catalysts or not) to produce low molecular weight intermediates that can be collected as oil and (2) subsequent solids formation via aromatization and polycondensation of the intermediates(Chen, Liu et al. 2020). In plastic carbonization, char (solid carbonaceous materials) is the main product, in comparison to the liquid product in fast pyrolysis and syngas product in gasification(Yaqoob, Noor et al. 2022). There are multiple carbonization pathways: anoxic pyrolysis carbonization, catalytic carbonization, and pressure carbonization (Figure 12).

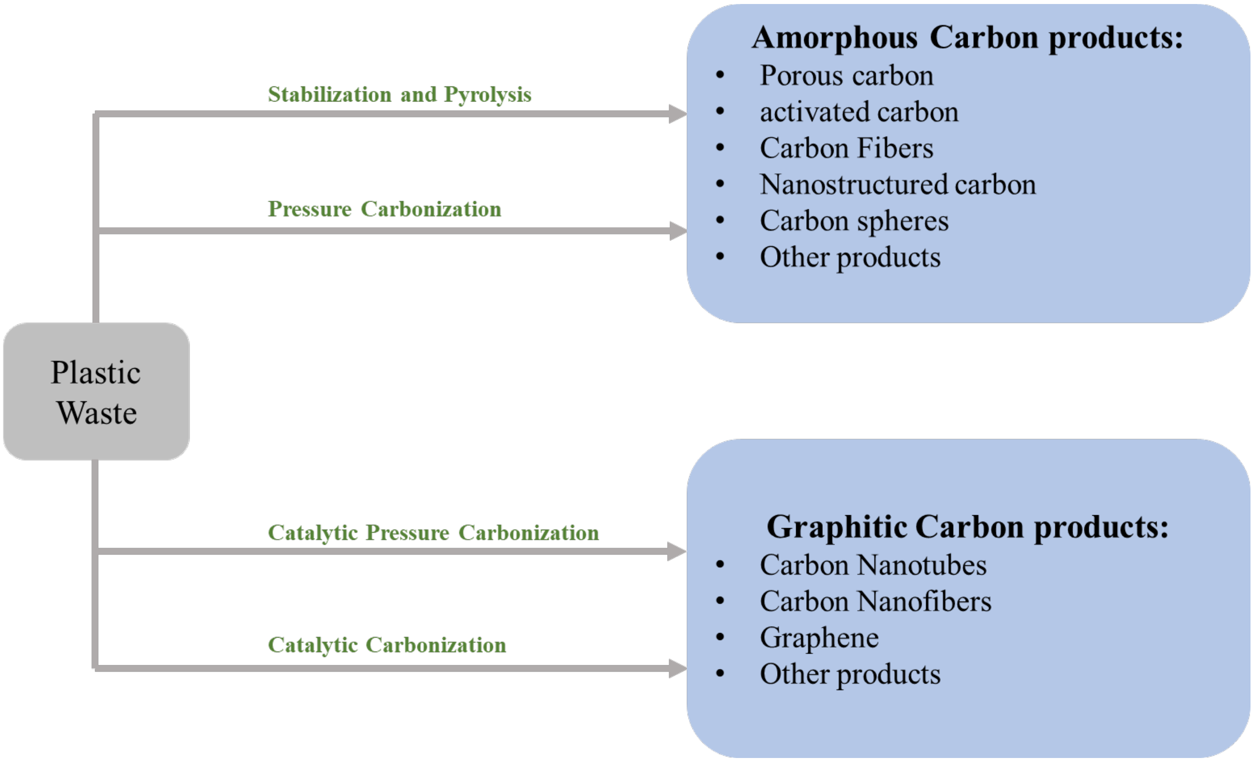

Figure 12. Schematic diagram of carbonization pathways of converting plastics or polymers to carbon materials (Adapted from Chen et al., 2020).

Plastic feedstocks for carbonization can be classified into charring (e.g., PF resin, PAN, and CPVC) and non-charring (e.g., PP, PE)(Gong, Chen et al. 2019). The backbone of charring polymers generally does not degrade but undergoes cyclization, aromatization, and crosslinking to form a carbon material frame(Chen, Liu et al. 2020). One challenge with a carbonization approach, which is still being addressed through research, is generally low yields. Higher carbon yields can be obtained in processing conditions under pressure (Geyer, Jambeck et al. 2017, Chen, Liu et al. 2020). Charring polymers (PF resins, PAN and CPVC) do not result in the formation of volatiles. (Li and Stoliarov 2014). Chars produced in carbonization processes can be activated to produce activated carbons(Merchant and Petrich 1993, Barranco, Lillo-Rodenas et al. 2010). In the case of non-charring polymers, it is critical to carbonize the volatiles to obtain advanced carbons (CNTs and/or CBs). To achieve a high yield of carbon (over 55%) from mixed plastics, it is necessary to control all the thermal cracking and carbonization steps. Several different processes are currently being explored for these purposes:

There are two different single-step carbonization strategies. Dry Carbonization includes technologies that take advantage of thermochemical reactions between 250 and 600 °C in a melted phase to obtain high char yields (Inagaki, Washiyama et al. 1988, Inagaki, Park et al. 2010, Fonseca, Meng et al. 2015, Akinyemi, Jiang et al. 2018, de Paula, de Castro et al. 2018, Barskov, Zappi et al. 2019, Castelo-Quibén, Pastrana-Martínez et al. 2019, Chen, Liu et al. 2020). Hydrothermal carbonization, a newer single-step strategy for carbonizing plastics, takes advantage of the same reactions as dry carbonization, but the presence of sub- and supercritical water enhances product removal and secondary reactions (Goto 2009). Condensation polymerization plastics (such as polyethylene terephthalate (PET), nylon, and polyurethane) and addition polymerization plastics (such as phenol resin, epoxy resin, and polyethylene) are relatively easily depolymerized to their monomers in supercritical water (Hu, Wang et al. 2010, Tran 2016, Dimitriadis and Bezergianni 2017, Helmer Pedersen and Conti 2017).

A third carbonization strategy occurs in two steps. This strategy involves decoupling the thermal degradation step with the production of an oil and a char, followed by the conversion of the oil into an advanced carbon (CNT or CB). The goal of this approach is to investigate and optimize the production of high-value advanced carbon materials from the volatiles.

There are several different products that can be created from carbonization. Carbon nanotubes (CNTs) are typically used in polymers, electronics, and lithium-ion batteries. The average market price is $600/kg (Dagle 2017, Khodabakhshi, Fulvio et al. 2020). The CNT market is estimated to grow from $3.95 billion in 2017 to $9.84 billion by 2023, reflecting a compounded annual growth rate of 16.7%). The multi-walled nanotubes represent approximately 92% of the total market. While most of the work on CNT production from plastics relies on hydrocarbon-based materials (PE, PP), a few studies have also shown success using oxygen-containing PET and PVA(Deng, You et al. 2016). Both plastics and biomass have been used to produce CNTs, but the products from plastic have better quality (Deng, You et al. 2016, Le and Yoon 2019). Because the synthesis of the CNTs depends on the nature of the pyrolytic vapors, it is important to advance our understanding on how to control the composition of these vapors using heterogeneous catalysts (Gong, Chen et al. 2019)

Carbon blacks (CBs), another possible product, are aggregates of nanoparticles of spheroidal shape, typically 10 to 100 nm in diameter, with an amorphous core surrounded by a shell of stacked graphene-like domains, each made of a nanometric stack of polyaromatic sheets (Khodabakhshi, Fulvio et al. 2020). CBs is the cheapest and largest-scale industrially produced nanocarbon with a projected annual production of 15 MT by 2025, suggesting the market would not easily be saturated, and a price $ 0.40-2.00/kg. The product is widely used as a pigment and as a reinforcing agent in rubber and plastics (Dagle 2017).

Cement and admixtures represent another product possibility. The North American market size for cement in 2018 was $12.3 billion (Anon). The global market for admixtures for building materials is estimated at approximately $15 billion (Plank 2004). With residential applications rapidly rising, the global tile grout and adhesive market, valued at approximately $2.2 billion in 2018, is expected to reach over $3.8 billion by 2026 (Chinchane 2019). CNTs, carbon macro, and microfibers have shown great promise in enhancing cracking resistance of cement systems(Siddique and Mehta 2014). Nanofibers can benefit cement composites by acting as extra nucleation sites, fillers, and pozzolanic materials, which can add to the first-crack strength and stiffness. Recycled CF reinforced polymers used as macro-size discrete reinforcements increase ductility and post-peak strength (Rangelov, Nassiri et al. 2016, Rodin, Rangelov et al. 2018, AlShareedah, Nassiri et al. 2019, Zhang, Lim et al. 2019). Calcium-silicate-hydrate is the most critical hydration product that contributes the most to the strength and durability of concrete/grout/mortar. Several key admixtures are added to facilitate the delivery of water and promote the formation of hydration products and, thus, the bonding capabilities in grout and mortar. A common admixture used in tiling grout is cellulose ethers, but this admixture requires costly equipment for the derivation process and is an expensive additive (relative cost of 200% compared to cement) (Plank 2004).

3.3.3.1 Summary of Key Messages

- In plastic carbonization, solid carbonaceous materials is the main product
- Carbonization Does not require complex pretreatments.
- Carbonization technology can produce a variety of carbon materials

3.3.4 Overall Key Messages for Chemical/Advanced Recycling

- Chemical/Advanced Recycling methods can handle heterogeneous plastic mixtures and contaminated feedstocks.

- A variety of products can be produced using Chemical/Advanced Recycling technologies.
- The ability of those processes to create higher-value products is one of the benefits of Chemical/Advanced Recycling technologies.

- Chemical or advanced recycling technologies fit into the circular economy by restoring the functional properties of plastic, providing added value to waste plastics, and investigating high-value applications.

4. Environmental Comparison of Plastic Waste Management Methods

landfilling and incineration are currently the most common methods for plastic waste management. A landfill can be used as a carbon sink for plastic waste if plastic doesn't leak into the environment. However, harmful materials in the plastic waste can be released into the soil and groundwater, leading to environmental concerns (Mahadevan Vaishnavi and Kannappan Panchamoorthy Gopinath 2023). Plastic waste has strong biological and chemical stability, meaning it does not decompose easily (Xiao, Yu et al. 2023). By using Landfill for the disposal of plastic waste, the plastic is lost from the value chain since they are not used as feedstocks for further applications. This does not support the circular economy and damages the environment(Davidson, Furlong et al. 2021). Moreover, landfilled plastic needs to be replaced by producing virgin plastic, which in turn is harmful to the environment since the process of fossil fuel extraction and virgin plastic production result in environmental issues.

The incineration of plastic wastes can be used to generate heat and electricity, which in turn reduces the need for fossil fuels and has the potential to decrease carbon emissions. However, in comparison with carbon-neutral technologies, the incineration of plastics results in an increase in emissions(Davidson, Furlong et al. 2021). Further, the emission of harmful gasses such as dioxins and furans resulting from the incineration of plastic waste needs to be accounted for and limits its use (Katami, Yasuhara et al. 2002). Moreover, the solids that remain after incineration contain a high amount of microplastics, which is harmful to the environment(Jiang, Shi et al. 2022). Incineration does not support the circular economy and leads to the loss of plastic from the value chain.

Life cycle assessment (LCA) is a tool to evaluate the environmental impacts of materials and processes throughout a product's life cycle (Finnveden and Potting 2014). LCA studies have indicated that any recycling technologies perform better compared to incineration and landfill, and Incineration performs better than landfill(Davidson, Furlong et al. 2021). Furthermore, the studies show that mechanical recycling performs better compared to chemical recycling despite the advantages of chemical recycling methods(Davidson, Furlong et al. 2021). Although mechanical recycling has a better environmental performance than chemical recycling, chemical recycling technologies have a wider range of application products with higher qualities.(Shen, Worrell et al. 2010).

Among chemical recycling methods, Pyrolysis is considered the most recommended method, although few studies have evaluated the environmental performance of chemical recycling methods, indicating that further studies are necessary to evaluate the environmental impact of chemical recycling techniques. (Davidson, Furlong et al. 2021)

5. Bio-based Plastic Recycling

Bioplastic can refer to plastic that is either biomass-based, biodegradable or has both properties (Merchan, Fischöder et al. 2022). Bioplastics have more sustainable plastic life cycles than fossil fuel-based plastic, and they have attracted much attention recently due to their lower environmental problems compared to fossil fuel-based plastic. Biodegradable bio-based plastics have shown a smaller carbon footprint compared with fossil-based plastics, and they have the ability to be compatible with existing recycling streams (Rosenboom, Langer et al. 2022). Therefore, they can be a preferred option in the circular economy framework.

It is important to note that plastics can be made from biobased materials but are not readily biodegradable (e.g., bio-based durable polyethylene). There are also some plastics, such as polybutylene succinate, that are typically fossil-fuel-based polymers, and they are biodegradable(Rosenboom, Langer et al. 2022).

Similar to the mechanical recycling of fossil fuel-based plastic, the mechanical recycling of bioplastics is associated with a reduction in bioplastic quality(Lamberti, Román-Ramírez et al. 2020). For example, the mechanical recycling of PLA resulted in a reduction in the mechanical performance of PLA (Żenkiewicz, Richert et al. 2009). The mechanical properties of PHB can be reduced as they are mechanically recycled(Rivas, Casarin et al. 2017).

A few literature studies exist on the chemical recycling of biomass-based polymers. Gasification and carbonization of bioplastic are in the early stages of research(Merchan, Fischöder et al. 2022). Pyrolysis has been used in several studies to add value to bioplastic waste(Sato, Furuhashi et al. 2001, Hirao, Nakatsuchi et al. 2010, Norrrahim, Ariffin et al. 2013, Arrieta, Fortunati et al. 2015, Li, Cai et al. 2020, Samorì, Parodi et al. 2021). For instance, The chemical recycling of PHA via pyrolysis resulted in the production of crotonic acid, which is a value-added material (Kumar, Sadeghi et al. 2023). The commercialization of chemical recycling processes of bioplastics remains limited, and technologies need to be developed since they are in the early stage of development, and more research is needed to evaluate the performance of chemical recycling processes.

Table 1. Summary of the advantages and disadvantages (or further improvements) of different routes for plastic waste management. (Patni et al., 2013, Gaurh and Pramanik 2018, Chua et al., 2019, Demetrious and Crossin 2019, Chen et al., 2020, Antelava et al., 2021, Li et al., 2022)

Method	**Advantages**	**Disadvantages or further improvements**
Landfilling	♦ Low cost in construction and operation. ♦ Easiest method for management	♦ The degradation of plastics takes an extremely long time. ♦ Takes up land resources. ♦ Releases plasticizers and produces microplastics, thus soil and groundwater pollution. ♦ Hinders the infiltration of surface water. ♦ Wastes plastic resources. ♦Contributes to a linear rather than circular economy.
Combustion or incineration	♦ Simple, low-cost, and efficient. ♦ Recovers some energy from the materials.	♦ Causes severe air pollution. ♦ GHG emissions. ♦ Generation of acidic substance and emission of acidic gases (e.g,. SO_x, HCl, NO_x) ♦ Contributes to a linear rather than circular economy,
Mechanical Recycling	♦Most common method for recovering plastics. ♦ Lower energy consumption than chemical recycling. ♦ Lower GHG emissions than chemical recycling.	♦ Not a suitable method for commingled and mixed waste plastic. ♦ The recycled plastics show relatively lower economic value compared to the initial plastics.
Carbonization	♦ Ability to control which carbon products are produced. ♦ Emission reduction. ♦ Can generate high-value products; ♦ Does not require complex pretreatments. ♦ Applicable for almost all plastic and biomass wastes.	♦ The practical application of carbon products needs to be further explored. ♦ Carbon recovery rates need to be further improved.
Pyrolysis	♦ High carbon efficiency ♦ Suitable for commingled and mixed waste plastic ♦ Variety of gas, liquid, and gas products	♦ Higher GHGs than mechanical recycling methods due to direct emissions and electricity consumption.

	♦ Suitable mixed waste plastic with biomass waste ♦ Lower emission of organic compounds compared to incineration	
Gasification	♦ Suitable for mixed waste plastic ♦ Suitable for co-gasification of plastic with biomass feedstock ♦ Wide range of products	♦ Higher GHGs than mechanical recycling methods due to direct emissions and electricity consumption.

6. Conclusions

This review discussed recycling techniques for plastic waste. The advantages and disadvantages of different routes for plastic waste management are summarized in Table 1. The most common recycling for plastics is mechanical recycling. Mechanical recycling is the current commercialized technique for the recovery of waste polymers. Mechanical recycling can, unfortunately, only treat a fraction of existing plastic wastes, and mechanical recycling of mixed and multi-layered plastics are challenging. Chemical recycling methods such as pyrolysis and gasification can handle heterogeneous plastic mixtures, which makes them a proper supplementary method for mechanical plastic. These technologies are capable of processing heterogeneous and contaminated plastic waste material, and they are suitable options where separation is not technically and economically viable. Although both gasification and pyrolysis can handle heterogeneous plastic mixtures, gasification has better flexibility regarding feedstock composition than pyrolysis. Moreover, a wide range of products can be obtained from the chemical recycling of plastics. One of the advantages of pyrolysis and gasification technologies is the ability to operate a mixture of biomass and plastic. In spite of advancements in gasification, pyrolysis, and carbonization technologies, commercialization of these technologies for plastic recycling confront challenges that can be addressed by more research and investigation. Moreover, the future market depends on policies, demand, and design. The policy can be a driving force to increase investment and collection throughout the recycling value chain. Finally, more chemical/advanced chemical recycling technologies are needed to be developed

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Appendices

Appendix A

Table A-1. Companies are working on developing chemical/Advanced recycling projects

Company	**Location**
Agilyx	Tigard, Oregon
Alterra	Akron, Ohio
Amsty	Woodlands, Texas
Anellotech	Pearl River, New York
Aquafil Carpet Recycling	Phoenix, Arizona
Braven Environmental	Zebulon, North Carolina
BiologiQ	Idaho Falls, Idaho
Brightmark	Ashley, Indiana
BASF	Port Arthur, Texas
Bioxyle	Atalanta, Georgia
Chevron Phillips	The Woodlands, Texas
Dupont Teijin films	Chester, Virginia
Dow Chemical	Midland, Michigan
Eastman	Tennessee
ExxonMobil	Bayview, Texas
Encina	Woodlands, Texas
Forell Pomini	Ansonia, Connecticut
Fulcrum BioEnergy	Pleasanton, California
Honeywell	Charlotte, North Carolina
Invista	North Carolina
Lummus	Houston, Texas
Milliken	Spartanburg, South Carolina
New Hope Energy	Tyler, Texas
Nexus Circular	Atlanta, Georgia
Nalco water	Naperville, Chicago
Novoloop	Menlo Park, California
PureCycle Technologies	Orlando, Florida
Protein Evolution	New Haven, Connecticut
Renewlogy	Salt Lake City, Utah
Regenyx	Tigard Oregon
Refined Plastics	Berks County, Pennsylvania
Scholle IPN	Northlake, Illinois
Shell	Park Norco, Louisiana
TotalEnergies	Tyler,Texas
LyondellBasell	Channelview, Texas
Loop Industries/ Indorama	Spartanburg, South Carolina
lanzatech	Skokie, Illinois

Appendix B

The feedstock should contain a minimum of 85% polyethylene or polypropylene**,** Maximum moisture content of 7%, and maximum total contamination of 15% (Adam Gendell 2022). Comparison of Model Pyrolysis Specification against Existing Sorting Specifications in Europe and North America are shown in Tables B1 and B2. Contaminants should not exceed the specific contents, including PVC(Polyvinyl chloride)/PVDC(Polyvinylidene chloride) (1%), PET(Polyethylene terephthalate)/EVOH(Ethylene vinyl alcohol)/Nylon (5%), PS(Polystyrene) (7%), Rigid metal/glass/dirt/fines(7%), Paper/organics(10%)(Adam Gendell 2022). However, both PE and PP are suitable feedstock for pyrolysis operators (while just polyethylene is usually of interest to mechanical recyclers) (Ahmad, Khan et al. 2015). PE and PP are the main desired feedstock of pyrolysis operators. Polyvinyl chloride (PVC) and polyvinylidene chloride (PVDC) films, introduce chlorine atoms into the pyrolysis process, which can lead to equipment corrosion and persist into the finished hydrocarbon product as heteroatoms(Bhaskar, Tanabe et al. 2005). Small amounts of PVC in mixed plastic are harmful due to generating HCl that corrodes the equipment (Lange 2021). One strategy to remove chloride could be to feed caustic elements such as $CaCO_3$ to the reactor to trap and neutralize chloride (Lange 2021).

The presence of oxygen atoms in the plastic waste feedstock generates oxygenated products, which causes yield reduction and negatively affects the quality of pyrolysis oil(Chang 2023). Breaking down the structures of complex hydrogen-carbons (e.g., nylon and PET) is not as easy as PE and PP, and the pyrolysis of these complex hydrogen-carbon produces impurities in the finished products(Adam Gendell 2022). Diluting the product with larger volumes of virgin hydrocarbons could be a strategy to use the product for lower-grade applications such as fuel or conduct hydrotreatment process, where hydrogen atoms are reacted with the product to combine with impurities, which facilitates their removal(Adam Gendell 2022). Ethylene vinyl alcohol (EVOH) and nylon have tighter limitations. Ethylene-vinyl acetate (EVA), ethylene-methyl acrylate (EVM), ethylene-acrylic acid (EAA), or polyurethane (PU) are expected to have similar challenges because their structure with nitrogen and oxygen functionalities. Paper and other organic materials which contain oxygen could be problematic, and hydrotreatment can be applied to remove these impurities(Belbessai, Azara et al. 2022). However, hydrotreatment increases the cost of the process. Studies show that pyrolysis can be profitable, and some factors, including feedstock cost, yield rate, product type, and facility scale, affect the profitability of the process(Li, Aguirre-Villegas et al. 2022).

Table B-1. Comparison of Model Pyrolysis Specification against Existing Sorting Specifications in Europe (Adam Gendell 2022)

	Pyrolysis Specification	**Plastic Films**	**Mixed Polyolefin**	**Flexible Polyolefin**	**Mixed Plastics**
Main composition	PP / PE	All polymers; sheet size >A4	Rigid and flexible PE and PP	Flexible PE and PP	PE, PP, PS, PET packaging
Min PE+PP content (min)	85%	Unknown	85%	90%	Unknown
Contamination (max)	15%	8%	15%	10%	10%
PVC/PVDC (max)	1%	Not stated	0.5%	Not stated	0.5%
PET/EVOH/nylon (max)	5%	Not stated	7.5%	5%	4% (clear bottles)
PS (max)	7%	Not stated	7.5%	0.8% (EPS)	Not stated
Rigid metal/glass/dirt/fines (max)	7%	0.5% metal 4% others	3%	1% metal 3% others	2% metal 3% others
Paper/organics (max)	10%		5%	3%	5%
Others (max)		Max. 4% rigid plastics	Undersize <20mm: max 2%		
Moisture (max)	7%	Not stated	Not stated	Not stated	Not stated

Table B-2. Comparison of Model Pyrolysis Specification against Existing Sorting Specifications in North America (Adam Gendell 2022)

	Pyrolysis Specification	**1-7 Bottles and All Rigid Plastic**	**PE Retail Bags and Film11**	**LDPE Colored Film**	**MRF Curbside Film**
Main composition	PO / PE	Rigid plastics	PE film	LDPE film	PE film
Min PE+PP content (min)	85%	unknown	95%	98%	95%
Contamination (max)	15%	5%	5%	2%	5%
PVC/PVDC (max)	1%		0%	0%	0%
PET/EVOH/nylon (max)	5%	Not stated	Not stated	Not stated	Not stated
PS (max)	7%	Not stated	Not stated	Not stated	Not stated
Rigid metal/glass/dirt/fines (max)	7%	1% metal	0%	0%	0%
Paper/organics (max)	10%	2%	5% 0% food waste	Not stated	Not stated
Others (max)		1% plastic bags, sheets, film	Other polymers, twine, and tape included in paper tolerance; 0% metallised films and multi-layer pouches	Max. 2% other polymers, labels and moisture; 0% metallised films and multi-layer pouches	Max 2% non-polyethylene other plastics, or labels
Moisture (max)	7%	1%	2%	Included in others	1%

Table B-3. Summary of waste plastic techno-economic analysis pyrolysis articles by feedstock, products, region, capacity, capital cost, and return on investment

Tech	Feed stock	Major products	Region	Capac (kton per year)	Capital Cost ($ mil)	Net present value (NPV) $	Ref
Pyrolysis	PS, PP, PE	Heavy oils petrochemical feedstock	United Kingdom	0.7–701	1.36–77.2	−0.44 per kg–0.71 per kg	(Fivga and Dimitriou 2018)
Pyrolysis + upgrading	PS, PP, PE, PET	Hydrocarb fuel	Korea	260	118	0.062 per gal	(Almohamadi, Alamoudi et al. 2021)
Pyrolysis	PE, PP, PET	Diesel power char	Australia	14.6	3.76	2.03 million	(Ghodrat, Abascall Alonso et al. 2019)
Pyrolysis & heat integration	HDPE	Ethylene propylene	United States	193	118.5 – 120.5	367.8 million–383million	(Gracida-Alvarez, Winjobi et al. 2019)
Fast pyrolysis	Mixed polyolefins mainly LDPE and residual PP	Naphtha	Belgium	120	Not disclosed	2.72-32.5 per ton	(Larrain, Van Passel et al. 2020)
Pyrolysis	Plastic waste (PP, PE, PS)	Light oil heavy oil	Malaysia	120	58.6	20.9 million	(Sahu, Mahalik et al. 2014)

Appendix C

Depending on the gasification method used, the quality of syngas is different. For instance, a syngas generated through air gasification of waste plastics has an average heating value of the 6–8 MJ m^{-3}(Lopez, Artetxe et al. 2018). Steam gasification could produce N_2-free syngas with a higher heating value(15 MJ m^{-3}). However, high tar is generated in gas products (Lopez, Artetxe et al. 2018). There are some technological challenges for gasification that need to be addressed. The low thermal conductivity of plastic waste, sticky behavior, high volatile content and tar formation can limit the application of conventional gasification technologies(Shah, Amin et al. 2023). The gasifier needs to be designed specifically for plastic wastes with specific features, including 1) the gasifier needs to have high heat transfer for fast plastic depolymerization, 2) it should be designed to handle the sticky nature of plastics, provide appropriate residence time for tar cracking and allow using catalyst in situ. Fluidized beds reactors have been wildly used for the gasification of plastic wastes(Mastellone, Zaccariello et al. 2010, Wilk and Hofbauer 2013). fixed beds, spouted beds and plasma reactors have been used for the gasification of palastic wastes(He, Xiao et al. 2009, Rutberg, Kuznetsov et al. 2013, Lee, Chung et al. 2014, Lopez, Erkiaga et al. 2015). The gasification process occurs through chemical reactions, which include drying, pyrolysis, cracking and reforming reactions in the gas phase, and heterogeneous char gasification(figure 9).

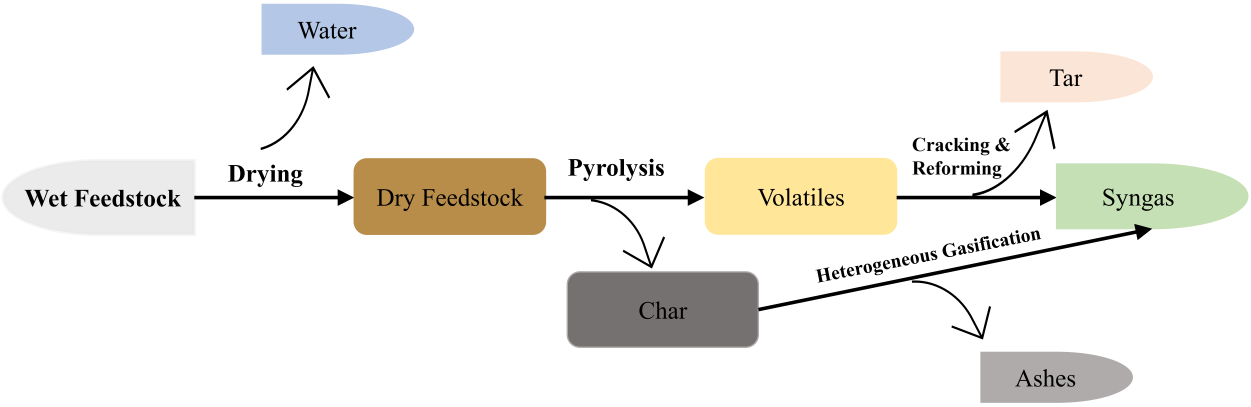


Figure C-1. Scheme of the main steps occurring in plastics gasification (adapted from (Rutberg, Kuznetsov et al. 2013)).

A series of chemical reactions of an endothermic nature occur in the pyrolysis step, which generates volatiles (gases and tars) and solid residue or char (Lopez, Artetxe et al. 2018). Under a fast heating rate in the pyrolysis step, common polymers such as polyolefins or PS can be converted into volatiles(Predel and Kaminsky 2000, Mastral, Esperanza et al. 2002, Anuar Sharuddin, Abnisa et al. 2016, Lopez, Artetxe et al. 2017). If the feedstock contains other materials, such as biomass, the char yield increases(Lopez, Artetxe et al. 2018). The amount of char produced is one of the differences between biomass and coal gasification and plastic gasification. Char gasification is a controlling step in biomass and coal gasification, which affects the gasifier design(Lopez, Artetxe et al. 2018).

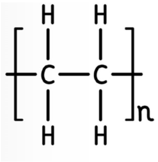
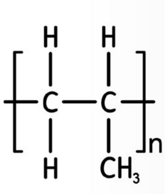
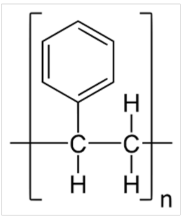
Catalysts are also used to improve conversion efficiency(Yang, Jan et al. 2022). For example, Ni-based reforming catalysts are active in the reforming of tar and other hydrocarbons and enhance H_2 Production(Lopez, Artetxe et al. 2018). Generally, more tar is formed in the gasification of plastics compared to biomass and coal(Li, Cañete Vela et al. 2021). Different gasifying agents(e.g., air, steam, and O_2) have been used for the gasification and co-gasification of plastics and biomass. The main challenge of plastic gasification for energy recovery is the presence of tar in gas products. The limit of the tar content is 10 mg Nm^{-3} for the use of the syngas for energy production in engines and turbines, and tar content must be even much lower than this limit for synthesis applications(Devi, Ptasinski et al. 2003, Lopez, Artetxe et al. 2018). Air gasification can be an option for energy production in the medium and small-scale power plants. The average lower heating value produced in waste plastic air gasification is in the range of 6–8 MJm^{-3}(Lopez, Artetxe et al. 2018). Plastic waste steam gasification can produce H_2-rich (3-18 wt. % of syngas) syngas with heating power to values above 15 MJ m^{-3} to be used for synthesis and energy applications (Lopez, Artetxe et al. 2018). Although the syngas obtained in plastics steam gasification has a higher quality, the endothermicity of the process and higher tar content in the gas product need to be considered (Lopez, Artetxe et al. 2018). Feedstock tipping fees, capital costs, and market prices are important factors in evaluating the profitability of the gasification process(Li, Aguirre-Villegas et al. 2022). Studies have indicated that the gasification of waste plastic is more profitable compared to conventional waste management practices such as incineration and landfilling(Di Gregorio and Zaccariello 2012, Bora, Wang et al. 2020, Voss, Lee et al. 2021, Li, Aguirre-Villegas et al. 2022).

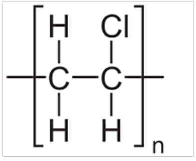
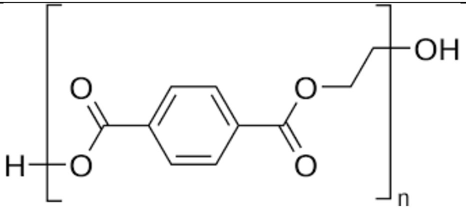
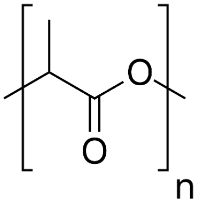
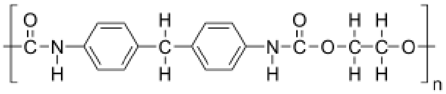
Appendix D

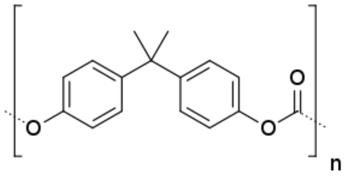
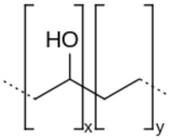
Common Types of Plastic in Waste

The most common plastic waste is summarized in Table D-1.

Table D-1. Common Types of Plastic in waste

Polymer	Structure
Polyethylene(PE): PE is one of the most extensively used and indestructible wastes, which is produced at a rate of 80 million tons per year(Guironnet and Peters 2020). PEs are produced under different operational conditions with different grades, chain branching, and density(Roy, Garnier et al. 2021). The higher crystallinity causes higher density, and branching leads to lower crystallinity (Roy, Garnier et al. 2021). PE with low density and crystallinity and high amorphous content is used for food packaging. High-density PE is usually used for packaging bottles for shampoo and detergents, gas pipes, water pipes, fuel tanks, and industrial wrapping films(Roy, Garnier et al. 2021). High-density PE is a safer option than PVC because it doesn't have chlorine(Kumar, Panda et al. 2011, Roy, Garnier et al. 2021). Various colors, additives, and adhesives are applied to produce PE-based products, which can negatively affect chemical recycling and upcycling processes (Vollmer, Jenks et al. 2020).	
Polypropylene(PP): PP is a thermoplastic polymer used in many products, such as the food packaging industry, electrical component manufacturing, reusable nonwoven bags, the automotive industry, nonwoven masks, and protective gowns(Roy, Garnier et al. 2021). PPs are produced around 55–56 million tons annually (Niu, Gonsales et al. 2019, Bora, Wang et al. 2020). One of the promising methods for the conversion of PP to value-added materials is to produce carbon nanoparticles and nanotubes via carbonization (Tessonnier and Su 2011, Roy, Garnier et al. 2021). Most products obtained from PP can be degraded by oxygen, light, heat, humidity, or shear forces during their lifecycle.	
Polystyrene (PS): PS is a thermoplastic polymer which is produced more than 20 million tons annually. PSs are usually used to produce cups, food containers, packaging materials, and insulators. Sorting Black PS-based food packaging waste by optical sorting system is challenging due to the high absorption of the black color (Hedayati, Barnett et al. 2019).	

Polyvinyl chloride(PVC): PVC is a thermoplastic polymer broadly applied in construction industries to make pipes, roofs, floors, doors, windows, and cable insulators. PVC is produced around 44.3 million tons annually, which is the second most wildly used plastic after polyolefins. PVC can contaminate the environment because of its high chlorine content; therefore, suitable waste management systems are essential(Roy, Garnier et al. 2021). Incineration of PVC also causes hydrogen chloride to be emitted into the air, which pollutes the air(Roy, Garnier et al. 2021). Chemical recycling of PVC is challenging since it may generate chlorinated hydrocarbons, which are harmful emissions(Cao, Yuan et al. 2016).	
Polyethylene terephthalate(PET): This polymer is a polyester thermoplastic usually used to produce polyester fibers in textiles, films, and plastic bottles. It was estimated that around 30 million tons of PET were produced in 2017(Roy, Garnier et al. 2021). In WA State, the generation of 91K tons of PET was estimated in 2017, which is about 43% of all rigid plastic packaging(Englund, Li et al. 2021). This increased if PET plastic film packaging and other non-packaging applications are considered(Englund, Li et al. 2021).	
Poly(lactic acid) (PLA): PLA is a bio-based aliphatic polyester which is comparable properties with petroleum-based polymers (PE and PS). PLA can be used to produce packaging films, foams, bottles, cups, and other injection moldable products. Fossil-based plastics are cheaper, which makes them more popular to use. PLA is biodegradable at temperatures around 58° C(Rudnik and Briassoulis 2011). Studies indicated that PLA is hydrolyzed in several decades at temperatures around 20° C, but at 50°C, it takes between 45-60 days(Tokiwa and Calabia 2006, Kolstad, Vink et al. 2012).	
Polyurethanes(PU): PUs have many applications, such as foams, coating, paints, and adhesives. It was estimated that around 17 million tons of PUs were produced annually(Roy, Garnier et al. 2021). The pyrolysis of PU may produce toxic gases such as hydrogen cyanide and ammonia(Roy, Garnier et al. 2021).	

Polycarbonate(PC): Poly(bisphenol A carbonate) is a synthetic polymer that is used to produce CDs, DVDs, airplane windows, water bottles, reusable food containers, bulletproof windows, safety glasses, travel suitcases, sports equipment, and electronic equipment(Roy, Garnier et al. 2021). The production rate of PC is around 6 million tons annually(Wang, Han et al. 2021). The incineration and disposition of landfill are harmful due to the release of bisphenol, which is a toxic compound(Samuilov, Korshunov et al. 2020, Wang, Jiang et al. 2020). Therefore, different strategies should be applied to manage.	
Ethylene-vinyl alcohol (EVOH): Most food packages need to be protected against oxygen. In package design, EVOH is used to block oxygen, it has a better efficiency than PE, PET, or nylon(Ragaert, Delva et al. 2017).	
Mixed Plastic: waste management of mixed plastic is highly challenging. Cleaning and sorting plastic waste are costly, and few proper commercialized technology have been recognized for recycling and upcycling mixed plastic waste (Roy, Garnier et al. 2021). Pyrolysis, gasification, and carbonization have shown the ability to produce high-value materials from mixed plastic. A commercial-scale waste plastic carbonization using a coke oven is used in Japan to produce coke, coke oven gas, and hydrocarbon oils from coal mixed with the waste plastics. The chlorine generated from plastic at high temperatures (1100° C) is trapped by the ammonia liquor that is used to cool the coke oven gas(Roy, Garnier et al. 2021). Thermochemical conversion technologies may produce a lot of volatile organic compounds, which makes the use of these technologies challenging(Roy, Garnier et al. 2021). Segregation, sorting, cleaning, and drying are the biggest challenges for the upcycling process.	