

9.1.2 Petrochemical Feedstock Recycling

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Against the backdrop of increasing production of packaging waste, an increased emphasis on the problem of microplastics in the marine environment, and the need for action in dealing with residual waste locally as a result of Chinese and other Asian import bans on waste, the EU Commission published the first EU plastics strategy in January 2018 [1]. In this strategy, 55% of plastic waste will have to be recycled in 2030, which will require the deployment of additional recycling capacity of ca. 10 Mt/a. The individual measures to be taken by the EU-27 countries are currently focussed on packaging waste [2]. A particular challenge is that the recycling rate of plastics will need to increase significantly to meet these targets. As a minimum prerequisite, suitable waste collection as well as sorting systems have to be implemented, and depending on the impact of these, additional technology pathways may be needed.

Clearly, waste recycling must be a pillar of the “circular economy” concept. A circular economy for plastics is hampered mainly by the nature of waste. Affordability is an issue, too. Plastic waste typically does not appear as a pure polymer. Recycling processes are either physical (meaning that materials are separated from each other but the molecular structure is not changed) or chemical (meaning that the molecular structures are changed and new materials are made from waste). Waste recycling requires the rigorous implementation of waste collection in general, but can be an effective means of recovering materials that can be re-used and recycled using standard, well-established processes. Not all materials can be recycled easily and at low cost, however, and in the absence of energy recovery, these are inevitably sent to landfill.

There are numerous barriers to the physical (mechanical) recycling of plastic waste, resulting in waste recycling rates that are below the targets mentioned above. These include: composite materials that cannot be separated mechanically, constraints associated with sorting technologies, non-melting thermosets and elastomers that cannot be re-granulated for second use, end-of-life products in complex mixtures and the associated high costs of separation, and ensuring the quality of secondary raw materials to maximize recycling. This results in the majority of plastic waste not being recycled; these include packaging waste sorting residues, shredder residues, thermal insulation composite systems, and waste from electric and electronic equipment. The recycling rate is higher for production waste than for post-consumer waste due often being single origin and having low levels of contamination. To date, post-consumer plastics waste in developed countries consists of equal proportions of packaging materials and engineering plastics. The size of the gap between current recycling rates and the targets varies from country

to country, in the same way as waste collection systems and application of (physical) sorting as a prerequisite for recycling do.

It is also difficult to remove the need for carbon entirely in many industrial processes. In a future independent of the use of fossil hydrocarbons, carbon will be available from biomass, anthropogenic wastes and residue streams, and from unavoidable CO₂-streams. Thus, the feedstocks utilized by the chemical industries in particular will change dramatically. Continuously increasing manufacturing of chemical products as well as high energy-density compounds (e.g. aviation fuels) will require these carbon feedstocks, and in order to meet climate protection targets. However, fuels are not covered in this chapter, as waste-based fuels are not regarded as waste recycling.

The aforementioned challenges and opportunities have increased interest in the potential of chemical recycling (also referred to as 'feedstock recycling' and 'waste-to-chemicals') to contribute to the recycling rate as an extension to waste sorting and as an alternative to waste incineration. Moreover, by reducing the fossil feedstock share in chemical manufacturing, chemical recycling of waste providing suitable chemical feedstock can reduce the carbon footprint of the manufacturing industry. A recent technology trend is a focus on chemical recycling of packaging waste to petrochemical feedstock, especially as a "drop-in" naphtha surrogate for multi-product steam cracking processes, accompanied by the "mass-balance approach" to the value chain, in order to recognize the contribution of waste-based virgin plastic to achieving the required recycling rates for plastics.

This section describes the basic principle of petrochemical feedstock recycling. It starts with an overview of plastics recycling processes according to their physical or chemical nature. Then, waste-to-petrochemicals process chains are discussed, linking plastic waste changes with the chemical feedstock requirements. Finally, key aspects are outlined.

Later in this book, Chapter 11 classifies thermochemical recycling processes and discusses the readiness, limits, and opportunities of the most important technologies for the case of mixed plastic waste, and gives guidance on process selection.

9.1.2.1 Waste & Recycling

Approximately 44% of global plastics production of ca. 390 Mt/a (including ca. 30 Mt/a of recyclates) is consumed by the packaging sector [3]. Worldwide, plastic waste generated amounts to ca. 250 Mt/a, of which nearly 57% is packaging plastics. The gap between production and waste and the high importance of packaging plastic waste is due to the short life-cycle of packaging versus the long life of engineering plastics products.

The situation is different for some developed countries with high gross national product that export technical products. Here, packaging amounts for only 25% of

plastics production, followed by the construction sector, automotive, and electrical and electronic products [4]. In these countries, packaging accounts for only about 40% of plastic waste, and often plastics recycling is already emphasized.

In both situations, a large amount of plastic is “stored” in plastic products in use, and this huge quantity, which has not been designed for recycling, grows continuously. This shows that (i) current focus on packaging waste recycling tackles the most important problem and can be a first step into a circular economy for plastics, and (ii) plastic products other than packaging, which are more difficult to recycle, will be of higher importance in the future.

Today, most plastic recyclates are sourced from separate collection of products consisting of relatively pure resins such as plastic bottles or manufacturing residues, or from sorting of collected post-consumer packaging waste by separating polyolefinic packaging products by highly developed sensor-based sorting technologies [4, 5]. The overwhelming amount of plastic waste that is not recycled is mixed plastic waste. Figure 9.1 illustrates the composition of mixed plastic waste for the example of a sorting residue from collected packaging waste. Besides different packaging plastics, this waste sample includes other packaging materials (e.g. metals and engineering plastics, but also biomass and mineral contamination). Many products included are composite materials, and all plastics included are functionalized, meaning that besides the pure resins they contain other plastic and non-plastic components, depending on the specific application they were used for. Achieving higher recycling rates means recycling this mixed plastic waste.

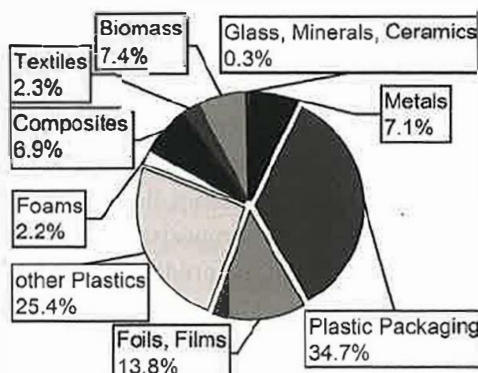


Figure 9.1 Macroscopic composition of a random sample of the lightweight packaging sorting residue as received, determined by a manual sorting analysis [6]

This section mainly emphasizes processes and technologies capable of processing mixed wastes at high temperature. The base thermal waste treatment process is waste incineration (“waste-to-energy”, WtE), i.e. combustion of waste with sufficient oxidant to allow full conversion of the hydrocarbon content to CO₂ and water.

For alternative thermal treatment (thermochemical conversion) which recycles waste to new feedstock, we distinguish between sub-stoichiometric oxidant availability (gasification) and processes with no additional oxidant feed (pyrolysis).

Gasification is an established process for production of chemicals and liquid fuels from coal; however, its use to manage waste streams is much less developed. The desired gasification product is syngas (primarily CO and H₂) of a specific composition depending on the downstream utilization process. Autothermal gasification in general delivers the necessary temperature level for a high syngas yield by direct heating, i.e. partial combustion of the feed, resulting in the byproducts CO₂ and H₂O. Like the one-step incineration process, gasification processes can be applied to generate energy in combined heat and power (CHP) plants. However, in contrast to incineration, gasification can also be set up as a two-stage combustion process, in which the feed is gasified first and the syngas is burned afterwards to generate heat. It is also possible to utilize the syngas as a secondary feedstock, which can be converted in downstream chemical synthesis to either “green fuels” or to basic chemical products – which demonstrates the flexibility of gasification pathways for waste management.

The definition of **pyrolysis** includes purely thermal cracking of the feedstock, catalytic cracking, and/or hydrocracking. In case of real mixed waste, it typically includes chemically bound oxygen, resulting in oxygen containing products such as water. Due to the complex degradation mechanism of plastics as well as biopolymers yielding a broad molecular product distribution, any pyrolysis process produces at least three multicomponent product phases, namely permanent gas, condensable vapours, and charcoal. Pyrolysis is an endothermic process that requires reaction heat supply through indirect heating of the reactor. It can be provided by means of combustion of part of the pyrolysis products. The purpose of pyrolysis is to convert biomass or mixed waste into secondary feedstocks for the production of fuels or chemicals. Although also being called “liquefaction”, due to the nature of the pyrolysis process in some cases, it cannot liquefy more than approximately 50 mass% of the feed, depending on its composition and the pyrolysis conditions. Furthermore, the multi-phase nature of the product stream can result in a need for additional downstream processes.

The scheme in Figure 9.2 organizes recycling processes according to their physical or chemical nature. Recovered components maintain their chemical nature during physical recycling processes (e.g. material recovery of a specific polymer), whereas chemical processes, in general, break up the initial material (e.g. recovery of the monomer as building block for new material).

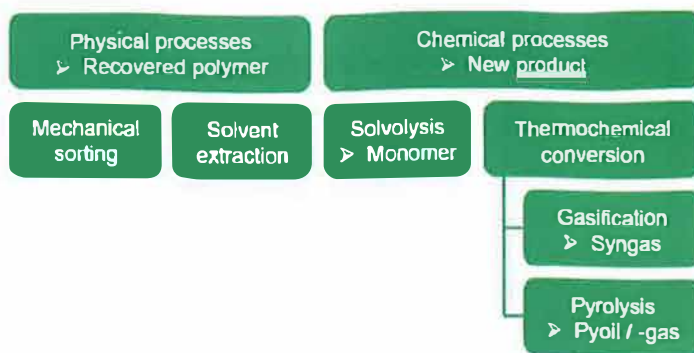


Figure 9.2 Recycling processes for plastic waste and respective main products

Physical separation processes (mechanical treatment, see Chapter 8) are well-established technologies to separate valuables from waste mixtures. Higher plastic recycling rates can be achieved by broad application of sensor-based sorting technologies, or in some cases by applying polymer-type-specific solvents. However, these technologies are characterized by high specific cost compared to basic mechanical waste pretreatment (such as the crushing, screening, etc. used for metals and minerals). The long-term increase of eco-products (“design for recycling”) will also lead to a higher rate of recovery in the packaging sector. Here the focus lies on physically separable thermoplastics. The polymer is recovered as so called recycle and can be processed into recycled plastic products. An obstacle is the lower quality of these mechanically recycled products compared to new virgin products. In general, re-granulates cannot achieve the quality of virgin material, because of (a) chemically bound ingredients such as ultra-fine particles (which can affect color) and catalyst residues, and (b) different mechanical properties resulting from the reduction in polymer chain length during post-processing.

Solvent extraction of one or more polymer types from plastic products also falls into the category of mechanical recycling. If applicable to a given mixture, this can result in clean polymer, thus overcoming the issue of quality. Initial applications on separately collected products are currently being run on a demonstration scale.

Solvolytic chemical separation processes are mainly applied to pure plastic waste streams available from plastic manufacturers or from plastic processing industries. Recently, solvolysis has focussed on suitable (pure resin) post-consumer products that are separately collected, such as plastic bottles and matrasses (see Section 9.1.1). This depolymerization through chemolysis is mostly possible with polycondensates. The monomers are recovered from the mixture and can be re-processed to high-quality virgin material. Key obstacles to increased adoption of this processing technology include contamination of the feedstock and the high process-specific separation effort.

Non-selective solvent liquefaction applying a hot, pressurized solvent aims to break down the waste polymer structures primarily into a “slurry” of liquid components. These reactions can use supercritical solvents, with an example being hydrothermal liquefaction of biomass. Like gasification and pyrolysis, it falls under the category of thermochemical conversion.

In general, **thermochemical processes** (thermal conversion, thermolysis) of waste decomposes mixtures of substances at high temperatures into the basic organic “building blocks” of chemistry, from which new products of high quality are produced. During processing, metallic and mineral impurities are separated and hazardous organic substances are destroyed. In principle, thermochemical processes are also suitable for those waste fractions that cannot be sorted by the above-mentioned processes, for example the non-pure fractions resulting from separate collections of packaging waste, such as sorting residues and shredder fractions. In general, mixed composite materials as well as plastics contaminated with catalyst residues or additives such as flame retardants can be recycled thermochemically.

In the following sections, chemical recycling of mixed plastic waste is considered. Herein, consecutive mechanical and chemical recycling go hand-in-hand in a waste-to-chemicals process chain, in which thermochemical recycling by pyrolysis or gasification avoids incineration of mechanical recycling sorting residues.

9.1.2.2 The Waste-to-Chemicals Process Chain

Taking the example of pyrolysis, Figure 9.3 shows the major feedstocks and intermediate products and the transforming process steps in the waste-to-chemicals process chain. Collected mixed plastic waste is first recycled mechanically. These processes are described in detail in Chapter 8. The products of these initial sorting steps are high-quality standard thermoplastics (which can be used as recyclates after washing and re-granulation), as well as residues. These residues typically include valuable materials like ferrous and non-ferrous metals, and also residues to be disposed of or treated, such as mineral fractions or waste water. Depending on the input waste stream and the mechanical pretreatment processes, a large combustible fraction remains as waste sorting residues. Its volume can be as high as 60–80% of the initial mixed waste [5]. Because modern political and legal boundary conditions do not allow landfilling of this fraction (see Chapter 5), the usage options are either energy recovery or chemical recycling.

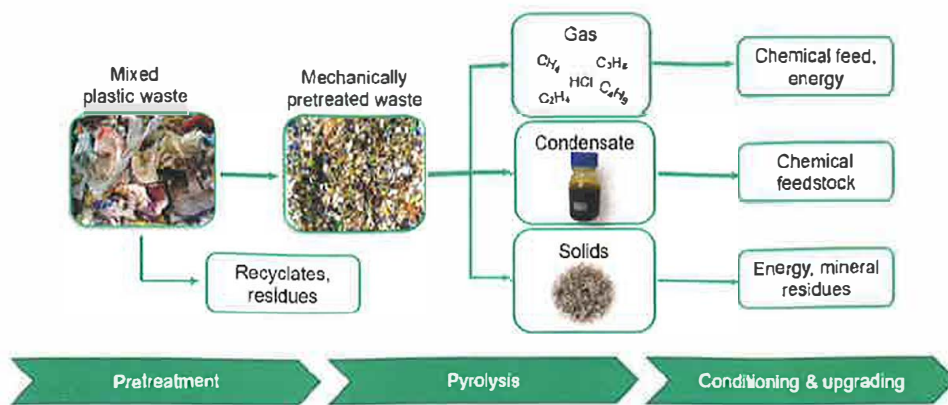


Figure 9.3 The waste-to-petrochemicals process chain: combined mechanical and chemical recycling using the example of pyrolysis

Energy recovery requires further conditioning of waste sorting residues into waste-derived fuels according to the requirements of the type of downstream energy recovery plant involved. These fuels can be specified as solid recovered fuel (SRF) if they meet the classification and specifications given in EN 15359 “SRF – Specification and classes”. An international standard for SRF, which is a more strictly defined sorted waste, is under development (ISO/TC 300 “Solid recovered fuels”). The most common options for using these fuels are in waste incineration plants for municipal solid waste, specific SRF plants, cement plants, and co-combustion in sewage sludge incinerators or coal fired boilers. Conditioning of SRF for this application includes mixing of mechanically pretreated waste fractions and adjustment of the fuel particle size distribution.

For recycling of waste sorting residues, similar conditioning to that for energy recovery would be necessary [7]. Here, we consider chemical recycling of this mechanically pretreated waste by pyrolysis. Processing of mixed waste by pyrolysis is described in more detail in Chapter 11.

Depending on waste feedstock, the pyrolysis technology applied, and desired downstream use, pyrolysis products require specific conditioning and upgrading:

- Pyrolysis solids include all minerals and metals separated from the waste (or added to the process to remove contaminants), as well as the pyrolysis char. This fraction typically needs to undergo thermal treatment, e.g. in a combustion step before further processing, and by this can contribute to deliver the pyrolysis process energy.
- Condensable pyrolysis products of feedstock derived from mixed plastic waste include a small water phase and a large oil phase. Pyrolysis water is to be treated for disposal, either through co-combustion with other pyrolysis product fractions, or through regular waste-water treatment. The pyrolysis oil itself, as the main product, is subsequently upgraded.

Non-condensable pyrolysis gas includes valuable gas species such as olefins as well as combustion products such as CO_2 . In particular, for pyrolysis of polyolefinic waste, the yield and value of pyrolysis gas can be so high that it becomes worthwhile to use it as feedstock, too. Otherwise, due to its relatively high heating value this fraction is incinerated, delivering process heat to fulfill the requirements of the pyrolysis process itself, as well as for supplying external heat or generating electrical energy.

9.1.2.3 Upgrading and Use of Pyrolysis Oil as Petrochemical Feedstock

The degradation of waste plastics into petrochemical feedstock paves the way to a closed plastics recycling loop, because when replacing fossil feedstock in polymer value chains, virgin-quality polymer product is generated from waste-based feedstock. By overcoming product quality issues of mechanical recycling and by replacing plastic waste incineration, chemical recycling can be a crucial step in increasing recycling rates, if the whole “circular economy” value chain performs better than fossil-feedstock-based polymer products in key areas such as climate impact and energy demand. The determinants of this are techno-economic feasibility and affordability. Recent studies have addressed the advantages of chemical recycling of packaging waste plastics by pyrolysis, including the impacts of avoided incineration [8–10].

By focusing on providing plastic-waste-based pyrolysis oils, large-scale existing chemical manufacturing infrastructures and value chains can be used, and extra investment is avoided. The waste-to-chemicals technical challenge is shifted to the plastic waste pyrolysis part of the value chain, and to the generation of suitable pyrolysis oils according to the specifications of downstream petrochemical processing units. The generation of waste-based pyrolysis oils and their composition are discussed in Chapter 11. The basic options for using pyrolysis oil feedstock are given in Figure 9.4. Here, key petrochemical value chains are ranked by their technology-specific feedstock quality requirements in terms of acceptable impurities:

- On the bottom level, partial oxidation of heavier hydrocarbon fuels by means of entrained flow gasification is a common process to provide synthesis gas for production of C1 chemical intermediates with an H/C ratio of 1. As a large-scale process, it is located in some petrol refineries as well as on large chemical production sites. In this application, waste-based pyrolysis oil replaces heavy fuel oil. Therefore, some sulfur and heavy metal content as well as higher viscosity (along with higher molecular weight) is more acceptable than for any other petrochemical value chain. Even tolerances of halogen species can be higher because of the implemented upstream syngas cleaning steps before use for chemical synthesis.
- The next quality level is fluidized catalytic cracking (FCC) of gasoil, which is the largest chemical reaction step in petrol refining. FCC is the worldwide most im-

portant source for C₄ olefins, and is nearly as important as a propylene source as naphtha steam cracking [11]. Pyrolysis oil as a surrogate for gasoil must be free of heteroatoms to a large extent. In contrast, hydrocarbon content can vary widely, including aliphatic species (and their isomers) and aromatic species. Molecular weight is significantly more limited than for heavy fuel oil. Also, olefin content is restricted. The hydrotreatment step in some petrol refineries is not considered here as a pyrolysis oil outlet, because it instead produces fuel components than chemical synthesis feedstock, and thus does not contribute to recycling.

- Naphtha steam cracking where plastic-waste-based pyrolysis oil replaces part of the fossil feedstock is the most direct way to close the plastic loop by chemical recycling. Naphtha steam cracking is the major source of small olefins. Similar restrictions on feedstock specification apply as for the FCC process. In particular, olefin content must be very low. In addition, boiling behavior, and more specifically start and end points of the boiling curve of waste pyrolysis oil, must be close to the feedstock design values for the specific steam cracker plant. Because pyrolysis oils are process-dependent multi-component mixtures, chemical pretreatment through adapted hydroprocessing is necessary to modify their product composition and properties before they can be blended with (or added to) light or heavy naphtha feedstock.
- Thermal separation of high value chemicals from waste-derived pyrolysis oils can be an option, if the content of specific olefinic or aromatic components in the multi-species mixture is exceptionally high. Therefore, the waste feedstock content of polymers that tend to form these intermediate species during pyrolysis must have been high (see Table 9.2) and pyrolysis parameters and technology must be adapted. The feasibility of thermal depolymerization with high monomer selectivity is addressed in Chapter 11.
- In general, waste based as well as biomass-based pyrolysis oil cannot fulfill all specifications of crude fossil-based oils. Due to its nature, the chemical composition and physical properties vary widely and cannot be adjusted except by hydrotreatment processes, if a narrow parameter variation and small deviations from petrochemical value chain feedstock specifications is required. This “heteroatom challenge” includes removal of halogens, removal or conversion of char- or water-forming precursors, and removal of components that can deactivate or poison catalysts in subsequent processes of the value chain. This challenge is partly overcome by blending pyrolysis oil with standard feedstock in these large-scale processes, as well as by choosing the petrochemical value chain that provides the best fit to the affordable pyrolysis gas quality. Due to the large size of the petrochemical value chains, it is reasonable to assume that blending capacity will be sufficient in the future, even with increasing volumes of recycled liquid feedstocks.

- A key issue for “climate-neutral” waste- and biomass-based feedstocks is that they have a lower hydrogen content than fossil feedstock (although plastic-waste-based feedstock is better than biomass-based feedstock, and both are far better than CO₂). Therefore, as the proportion of these feedstocks used in chemical production is increased, so does the amount of (renewable) hydrogen required. The highest chemical recycling efficiency will be achieved when plastic-waste-based feedstocks are converted to products with a similar C/H ratio.

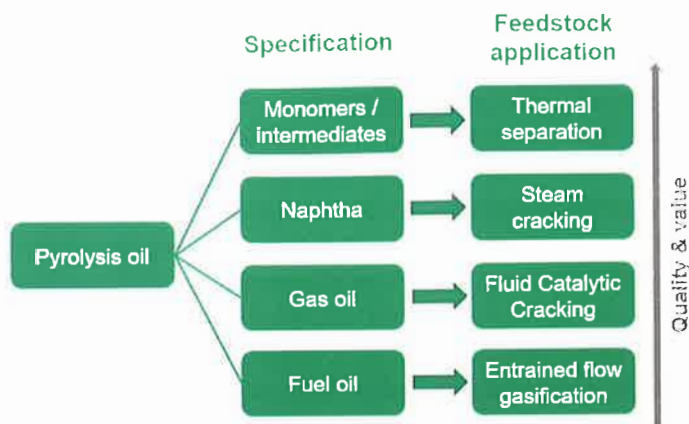


Figure 9.4 Petrochemical feedstock recycling options through pyrolysis oil – specifications to be achieved by the entire pyrolysis process according to feedstock application

9.1.2.4 Decentralized Versus Centralized Chemical Recycling

Chemical recycling plants can be decentralized, i.e. located at the primary (mechanical) waste treatment plant where collected waste is made available for further processing, or centralized, i.e. at the petrochemical production site where the waste-based feedstock is processed.

The overall efficiency of the waste-to-chemical process chain can depend strongly on this choice in terms of both the environment and the economy. Two key aspects influence this decision – transport effort and yield of the chemical recycling product. Whilst waste is typically lower in density and energy content than a liquid chemical recycling product that is determined as feedstock, it is nevertheless typically collected and transported over long distances. Specifically in urban and industrial regions, transport effort will thus not be a major consideration on decentralized versus centralized plant location. This is not the case if plastic foam is being recycled, when transport effort can be a deciding factor on the location.

If a key product of a chemical recycling process is gaseous, decentralized recycling is not favored. At decentralized locations, gaseous products cannot typically be utilized as feedstock and their value must be recovered energetically. Combustion

of the gases brings little economic and environmental value. Thus, gasification processes, as well as pyrolysis processes with a high yield of gaseous products compared to pyrolysis oil, benefit from a central location and integration into petrochemical sites [12]. Pyrolysis of mixed packaging plastic waste is one such case. Here, on the one hand, the oil yield of thermal pyrolysis can be limited due to the heterogeneous waste mixture, because it depends highly on the cleanness and preselection of the waste feedstock, i.e. a high content of polyolefins. If catalytic pyrolysis is applied to such a feedstock, the yield can dramatically shift from the oil to the gas phase. Furthermore, the quality of the pyrolysis gas can be drastically increased. Therefore, central pyrolysis at a steam cracker site would be preferable, where gases as well as oils are utilized as feedstock, and where upgrading of pyrolysis oil can be done centrally.

9.1.2.5 The Status of Petrochemical Feedstock Recycling

The present trend towards chemical recycling is steam cracker feedstock recycling [13] of packaging plastic waste by means of pyrolysis. Packaging waste is the largest and most recognized waste stream, pyrolysis of which is clearly easier than converting engineering plastics to chemical feedstock, while also allowing design of demonstration plants and subsequent developments with a higher technology readiness level (TRL). Focusing on decentralized provision of pyrolysis oils initially avoids larger investments and integrates into existing petrochemical value chains with a focus on plastics-to-olefins recycling. These technologies and their current applications are given in Section 9.1.1.

By leading to new products, thermochemical recycling of mixed plastic waste has the potential to overcome mechanical recycling issues, as well as problems from the relative purity of waste feedstocks when used in solvent-based processes. Therefore, it is worth significantly increasing the technology readiness level of pyrolysis and gasification processes by improving feedstock flexibility, with a focus on removal of contaminants and heteroatoms from liquid products. Furthermore, these processes address recycling of composite materials and non-melting plastics. They can therefore complement mechanical and chemical recycling process chains towards the goal of achieving higher recycling rates and economic feasibility, helping chemical recycling compete with waste incineration.

Thermochemical processing of mixed plastic waste is the scope of R&D efforts. Because so far there are only a few large-scale applications for selected packaging waste only, data availability on chemical recycling processes that allow for process evaluation is very limited. Laboratory data cannot be scaled-up because mixed feedstocks as well as applied technology have a critical impact on product yields and compositions. Chapter 11 will address mass and energy balances, yields, and application examples of pyrolysis and gasification technologies.

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