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Chemical recycling

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Section III 2.4, III 1.6, III 1.2
PO Box 14 06
D-06813 Dessau-Roßlau
Tel: +49 340-2103-0
buergerservice@uba.de
www.umweltbundesamt.de

 [/umweltbundesamt.de](https://www.facebook.com/umweltbundesamt.de)
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Authors:

Dr. Julia Vogel
Dr. Franziska Krüger
Matthias Fabian

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1. Background

Plastics are an important part of our lives and our economy, present in all areas of life and often difficult to replace. However, plastics that become waste must be treated appropriately and recycled to the highest possible standard. The waste hierarchy of the EU Waste Framework Directive 2008/98/EC ([1]) defines a general order of priorities for waste management measures, according to which recycling is considered to be fundamentally more advantageous than energy recovery (Article 4 Waste Framework Directive) [2].

Recycling is not an end in itself, however. It must meet the requirement of providing the best possible protection for people and the environment, taking into account the entire life cycle of the waste. Plastics recycling should be carried out to the highest possible standard with the aim of recovering materials from the waste stream that replace primary materials and, ideally, primary plastics, thereby conserving resources (through the process known as material recycling). Moreover, the recyclates produced from plastic waste should be used in appropriate and purposeful applications.

The German Environment Agency defines plastics as solid materials that consist mainly of synthetically produced or chemically or biologically modified natural macromolecules or polymers. Plastics also contain other substances such as additives (e.g. antioxidants, processing aids) or fillers (e.g. lime, glass fibers), which are added to optimise the functional properties of the various polymers. Liquid polymers do not fall under this definition.

However, recycling can only be performed on recyclable materials¹ for which recycling technology is available. In particular, the wide range of materials falling under the category of plastics [3] creates much more difficult recycling conditions than in the case of paper or glass, for example. Product design, which is often not geared towards the needs of recycling, is also regarded as an obstacle to mechanical recycling, in which the structure of the plastic polymers and the material remain largely preserved. Furthermore, durable plastic products in particular may contain production-specific ingredients that have now been identified and regulated as harmful substances (e.g. insulation boards with certain flame retardants). If substances restricted in their use cannot be specifically removed or the waste cannot be managed in a safe closed cycle, it must instead be used for energy recovery or even disposed of in thermal disposal facilities.

In recent years, plastics have received a great deal of media attention and have thus become the focus of public interest. In this context, in addition to material recycling, the use of chemical or feedstock recycling (see section 2.2) is being discussed as another form of recycling that is considered to offer both the possibility of removing pollutants and the potential of utilising waste that is difficult to recycle or heavily polluted.

In January 2018, the EU Commission presented its plastics strategy [4] with the aim of regulating and improving the sustainable usage of plastics. Among other things, it defines strategies for the prevention of plastic waste, the improvement of product design in terms of increased recyclability, and the separate collection of plastic waste. These political objectives are implemented in concrete form in the Waste Framework Directive and the EU Packaging Directive 1994/62/EC (Packaging Directive [5]), among others. These regulations include recycling targets: the EU Packaging Directive sets high standards for the recycling targets to be met for plastics recycling

¹ For packaging subject to system participation requirements, the Central Agency Packaging Register and the German Environment Agency publish an updated minimum standard for determining recyclability, see: <https://www.verpackungsregister.org/en/foundation-authority/minimum-standard-pursuant-to-section-21-verpackg>

from 2025 onwards (Art. 6 para. 1 (g), (h) Packaging Directive). In Germany, this is implemented in the German Packaging Act [6] (*German: Verpackungsgesetz, VerpackG*, abbreviated from herein after as Packaging Act), which has set new, ambitious targets for recycling packaging materials since it entered into force on 1 January 2019 (section 16 para. 2, 4 Packaging Act).

In addition, Art. 11 para. 2 Waste Framework Directive sets ambitious targets for the recycling of municipal waste (50 % from 2020), which are to be incorporated into national law in Germany through amendment of the German Circular Economy Act [7] (*German: Kreislaufwirtschaftsgesetz, KrWG*; abbreviated from herein after as Circular Economy Act) (in section 14 para. 2 Circular Economy Act [7]).

However, this recycling target does not relate directly to the proportion of plastics contained in municipal waste, but to the total amount of municipal waste. In this context, chemical recycling is also being discussed as a possibility for recycling contaminated, mixed-plastic waste fractions from residual waste as well as residues from sorting plants that have so far been used for energy recovery [8].

According to the legal requirements, significant increases must be achieved in the amount of plastic waste collected and fed into recycling. This means that it will be necessary in the future to consider other plastic-waste streams that have not yet been recycled, as well as new techniques like chemical recycling, in order to increase recycling rates.

2. Techniques for plastics recycling

According to Art. 3 para. 17 Waste Framework Directive (transposed into section 3 para. 25 Circular Economy Act [9]), recycling refers to any recovery process by which waste is processed into products, materials or substances that are to be used either for their original purpose or for other purposes. This includes the reprocessing of organic materials, but does not include energy recovery and the reprocessing into materials that are intended for use as fuel or for backfilling. (Material) recycling encompasses mechanical and chemical or feedstock recycling processes. Mechanical recycling is sometimes also referred to as physical recycling and includes dry and wet mechanical recycling as well as solvent based recycling. However, in this paper the term mechanical recycling is used to refer to both mechanical and solvent-based recycling.

2.1 Mechanical recycling

In the recycling of plastic waste, mechanical recycling is the preferred method at present. This form of recycling involves processes in which the polymer structure is not significantly changed and the plastic is preserved as a material. Thus, in terms of its complexity, mechanical recycling takes place at a lower technical level than chemical recycling. This

is also associated with significantly less preparation effort, although it does necessitate appropriate qualities in the input materials.

The basic prerequisite for successful mechanical recycling is the most thorough possible sorting of individual plastic types in order to ensure a high degree of quality in the recovered secondary plastics. In practice, this is achieved through dry and wet mechanical processing methods or by extracting certain polymers using selectively active solvents.

2.1.1 Dry and wet mechanical processes

The combination of dry and wet mechanical treatment processes is designed to remove impurities (e. g. paper, metals, glass, minerals, non-targeted plastics) and to produce a plastic product with specific defined properties.

In *dry mechanical* processes, the plastics are separated from impurities using inductive and magnetic metal separation, air separation and sifting, and are then shredded. Sorting of the individual plastic types is usually carried out by the method of near infrared spectroscopy (NIR sorting). Alternatively, electrostatic separation is also used, e. g. for carbon black coloured- plastics [10].

The subsequent *wet mechanical* processing consists of shredding and multi-stage washing and separation processes. Washing serves to remove adhesions (e. g. food residues, cosmetic components, sand) as well as labels and adhesive residues. The targeted plastic types are recovered by means of density separation. Plastic types with different specific densities are separated from each other in a wet medium. Swim-sink basins, hydrocyclones and sorting centrifuges are the state of the art here. Plastics with very similar and overlapping density ranges require further sorting methods as part of dry mechanical processing, mostly by means of NIR technology or electrostatic separation.

The wet mechanical process is followed by drying of the plastic material through mechanical and/or thermal processes. Mechanical drying involves the removal of moisture by dry centrifuges. In thermal drying, by contrast, heated air flows through the moist material.

In addition to sorting by plastic types, aggregates for colour sorting are also sometimes used. This extends the colour spectrum of the recyclates and thus also the range of applications for which they may be used subsequently [10].

At the end of the material preparation process, the dried plastic regrind is either supplied in the form of flakes or processed into agglomerates or regranulates in a further processing step, depending on the requirements of the customer. In the agglomeration process, the plastic particles (regrind) are joined together by surface melting in order to achieve uniform particle sizes and higher bulk densities. To obtain homogeneous and free-flowing regranulates, the regrind gets completely melted under pressure and temperature in an extruder. Melt filters ensure the removal of any remaining impurities (e. g. wood fibres, plastics with higher melting temperatures), while volatile components are removed in a degassing unit by means of negative pressure. On exiting the extruder, the melt is pressed through a die plate, granulated and cooled down [10].

2.1.2 Solvent-based processes

In solvent-based methods, plastics are recovered in a physical process, in which the polymer structure is retained. With the help of specially adapted solvents, plastics are (selectively) put into solution. This is followed by a multi-stage process to separate liquid and solid components. In this way, it is possible to remove non-targeted plastics, additives and any pollutants that may be present. The target plastic is then precipitated, cleansed of solvent and dried. Finally, the recovered plastics are extruded into regranulates. The advantage of solvent-based processes is the possibility of recovering very pure plastics from composites or plastic waste containing harmful substances. However, these processes are either not available in Germany or only to a limited extent [11, 12].

2.2 Chemical or feedstock recycling

Chemical or feedstock recycling refers to the conversion of plastic polymers into their monomers or chemical building blocks or basic chemicals, i. e. depolymerisation by means of thermochemical or chemical processes, although there is currently no uniform, legally binding definition [13].

Chemical recycling is considered to have the potential to enhance material recycling by tapping into new types of waste that could not be recovered through mechanical means, thus making it easier to reach some of the targets for plastics recycling required by law [8, 14].

Possible chemical recycling techniques include pyrolysis, gasification and liquefaction (e. g. liquefaction in an oil bath or solvolysis). The use of plastic waste as a reducing agent in the blast furnace process, for example, is also considered part of feedstock recycling. However, the latter will not be examined further in the following, as no new input materials can be provided for plastics production, in contrast to the other processes described. In addition to the direct use of chemical recycling products as input material for the chemical industry, recycling to liquid energy sources/fuels is also considered part of feedstock recycling in some cases. This method is also excluded from further examination here, as it has no clear ecological advantages, from an environmental point of view, over direct thermal or energy recovery in waste incineration plants, substitute fuel power plants or cement kilns (referred to in the following

simply as energy recovery methods), especially when there is a need for products of high quality, as required for the production of fuels.

The various chemical recycling processes (pyrolysis, gasification, liquefaction in an oil bath, solvolysis) yield completely different products. Solvolysis breaks down the polymer chains into monomers, whereas liquefaction in an oil bath and pyrolysis produce hydrocarbon mixtures of different compositions which, after fractionation and processing, can then be used as feedstock in the chemical industry for example. Gasification technologies, in contrast, produce a synthesis gas (syngas), which in turn is converted into hydrocarbons in several subsequent steps (e.g. Fischer-Tropsch synthesis) and can thus also serve as a basic material for the industry.

The possibility of producing basic feedstock materials for the chemical industry makes the newly emerging processes of chemical recycling potentially attractive, as they could contribute to closing the gap between waste management and raw materials industries [8, 15]. Use of the carbon contained in plastic waste as a secondary carbon source opens up the prospect of effectively recycling it and thus contributing to decarbonisation of the industry, i.e. to discontinuing the use of fossil carbon sources, or at least reducing them, and thus significantly cutting greenhouse gas emissions. However, the technical suitability and the ecological and economic advantages of chemical recycling processes have not yet been conclusively demonstrated. Although some processes have already been tested, there has been no large-scale, sustained implementation to date.

In the following, the various possible technical approaches for chemical recycling are described in brief and general terms. For all processes, the waste input has a strong influence on the product quality obtained: the higher the degree of contamination and heterogeneity of the waste input, the greater the effort required to process the products and the greater the amount of residual materials that may need to be disposed of.

2.2.1 Pyrolysis

Pyrolysis is a process that has been in use for centuries, for example in charcoal production and the coking of hard coal. In this process, organic materials are thermally broken down in an oxygen-free environment [16, 17]. Depending on the input material used, this results in the formation of gaseous, liquid or solid reaction products [18, 19].

The use of mixed municipal waste in pyrolysis has not gained acceptance due to the heterogeneity of the input materials and the resulting poor quality of the products obtained at the end of the process. The pyrolysis of plastic waste with the aim of recycling is more promising when more homogeneous plastic fractions are used [16]; the extent to which more polluted mixed-plastic fractions can be used has not yet been conclusively determined.

The products of pyrolysis are oils and waxes, which can be used as basic materials in the chemical industry after further purification steps.

2.2.2 Gasification

Gasification processes treat organic materials thermally in reduced oxygen conditions and are well-established processes in the field of coal gasification, for example. Coal gasification was long used in Europe to produce town gas, for instance for the purpose of street lighting. Gasification can be performed through various techniques that remain common in China and other countries, and are still used in Europe, as well. However, these techniques have not been found suitable for the treatment of waste, especially municipal waste, although this has been attempted several times in recent decades [16].

The aim of gasification processes is to turn solid (and sometimes also liquid) materials, including those from plastic waste, into a syngas that can then be put to high-grade use. In the chemical industry, syngas (a hydrogen and carbon monoxide mixture) can be used as a basic material for the production of many chemical products. However, the composition and thus the quality of the syngas produced is highly dependent on the feedstock. In general, the more heterogeneous the feedstock, the more complex the purification processes required to produce a high-quality syngas.

2.2.3 Liquefaction

Liquefaction processes rely on very different techniques but all aim at the direct liquefaction of plastic waste and subsequent use of the liquid product, which usually has to be purified beforehand [19]. Processes that may be used for this include solvolysis and liquefaction in an oil [18, 20].

Solvolysis refers to the dissolution of polymers in organic solvents, if necessary under increased temperature and pressure [20]. In contrast to solvent-based processes, which preserve the molecular structure of the polymers, solvolysis breaks them down into their basic building blocks (monomers) [13].

Liquefaction in an oil bath refers to the direct thermal or catalytic decomposition of plastic polymers in a stirred tank reactor [19]. The resulting products are liquid, gaseous and solid. The target product represents the liquid phase, while the wax phase constitutes the solid residues, such as fillers and contaminants from the feedstock. However, depending on the input material, impurities can also be found in the liquid phase, and these must be removed before the product can be used in a high-grade application [16, 19, 20].

3. Evaluation of chemical recycling

The processes of chemical or feedstock recycling turn waste into products, materials or substances to be used either for their original purpose or for another purpose. Although there is no uniform legal definition of chemical recycling [13], it may be classified as recycling as defined by the Circular Economy Act and Waste Framework Directive and thus, according to the waste hierarchy, is generally considered preferable to energy recovery. In the general technical understanding, chemical recycling encompasses all processes for the depolymerisation of plastics. After treatment, the resulting products can be used as feedstock material for the chemical industry/plastics production.

In principle, it is also conceivable that chemical recycling products might be used as fuel. Such an approach, however, runs counter to the very concept of material recycling and therefore practically equates to energy recovery. Of the various methods available for recovering the energy content of plastic waste that cannot be recycled using the current technology, direct energy recovery is considered the state of the art, not chemical recycling. New processes must therefore compete with the two established process paths (mechanical recycling and energy recovery) and demonstrate their ecological advantages and economic feasibility. This has not yet been done.

3.1 Evaluation of techniques and input materials

From an ecological point of view, chemical recycling is particularly well-suited for plastic waste that is difficult or impossible to recycle using common techniques (e. g. sorting and processing residues, mixed plastic waste). Chemical recycling is technically possible in principle for the recycling of plastics, although this is highly dependent on the process selected, as a function of the input material. Plastic waste that is not yet recyclable via mechanical recycling could potentially be recovered through chemical recycling processes. In the view of the German Environment Agency, however, the material flows that have so far been used in mechanical recycling should not be redirected towards chemical recycling, as it can be assumed that mechanical processes, which are technically far less complex, are more advantageous in ecological terms at this stage.

High demands are placed on the chemical recycling process. In particular, pyrolysis and gasification, which have already been tested in other areas, are complex technical processes, for which the technical requirements and the quality of the products obtained are highly dependent on the contamination and composition of the waste used. Therefore, as in mechanical recycling, the pre-treatment of waste input material is of great importance. The greater the heterogeneity of the plastic waste used, the more

complex the process management and technical requirements involved. Process control, material and energy management, and downstream treatment are decisive for the quality of the products obtained, which in turn determines how useful and marketable they are. The assumption here is that plastic waste with significantly inferior qualities, a high level of heterogeneity and a higher degree of contamination than is suitable for mechanical recycling can pass through chemical recycling and thus be made useful. But this has not yet been conclusively demonstrated, and there are no large-scale, permanently operating plants on the market under conditions such as those required in Germany. Moreover, the operation of a large-scale pyrolysis or gasification plant, which is associated with high investment and operating costs, requires a high throughput in order to be cost-effective. This means that only plastic waste that is available on the market in large quantities and does not directly compete with mechanical recycling can be considered as input material.

The real viability of the various processes for chemical plastics recycling described above has yet to be demonstrated in large-scale implementation, although the processes as such have already been tested for other input materials. In particular, it is not yet clear which techniques are most appropriate and which plastic waste fractions can be recycled most effectively using which technology. Apart from the matter of technical feasibility, it is also unclear, so far, whether or not chemical recycling has ecological advantages in comparison to direct energy recovery from plastic waste fractions that cannot be subjected to mechanical recycling, taking into account all processing and post-processing steps; this remains to be evaluated in long-term operation [21]. No definitive conclusion can be drawn from the data currently available. A systematic analysis of the existing approaches is needed for this purpose. The German Environment Agency is currently carrying out a study in order to answer these questions (“Abschätzung der Potenziale und Bewertung der Techniken des thermochemischen Kunststoffrecycling – Assessment of the possibilities and evaluation of the techniques of thermochemical plastics recycling”, FKZ 3720343020).

3.2 Possible contribution to meet recovery targets set in Circular Economy regulations

Targets, usually in the form of input targets, are established for various types of recovery performed in Germany. They generally apply for

- ▶ recycling (section 3 para. 25 Circular Economy Act),
- ▶ mechanical recovery (section 3 para. 19 Packaging Act) or
- ▶ recovery (section 3 para. 23 Circular Economy Act), i. e. preparing for re-use, recycling and other purposes, in particular energy recovery, which together can contribute to meeting the target.

Some older legal norms make reference to *stoffliche Verwertung*, that is “material recovery”, (e. g. in the Batteries Act [22] and the End-of-Life Vehicles Ordinance [23]), with the definitions corresponding to those of “recycling” in the respective EU directives (End-of-Life Vehicles Directive 2000/53/EC, Batteries Directive 2006/66/EC). They differ, however, from the definition of “recycling” in the Waste Framework Directive 2008/98/EC, as, among other things, back-filling is not excluded, in contrast to the definition of “recycling” in the Waste Framework Directive. There is also a difference as to whether such specifications for recovery targets refer to the total weight of the waste (as stipulated, for example, in the End-of-Life Vehicles Ordinance and the Electrical and Electronic Equipment Act [23, 24]), or whether targets are defined in a material-specific way (as is essentially the case in the Packaging Act, for example).

In Germany, mechanical recycling targets specifically for the material plastics exist only for packaging waste. In the Packaging Act, which has been in force since 1 January 2019, the requirements for the introduction of (mechanical) recycling have been significantly increased compared to those of the packaging ordinance (*German: Verpackungsverordnung* [25]) that applied previously. Plastic packaging that is collected from private end consumers and similar sources via yellow waste bins/bags is subject to ambitious targets: the systems (producer responsibility organisations) are obligated to mechanically recover an annual average of 58.5 percent of their plastic packaging; with effect from 1 January 2022,

this target will rise to 63 % (section 16 para. 2 sentence 3 Packaging Act). Mechanical recovery involves processes by which new material is replaced by similar material or the material remains available for further material use (section 3 para. 19 Packaging Act). The recovery target for plastics packaging, i. e. the target for recycling, preparing for re-use and other recovery, including energy recovery, has been increased to 90 percent by weight of the plastic packaging quantity participated with the system (section 16 para. 2 sentence 2 Packaging Act). For other composites, including composites with a plastics content, there is a recycling input target of 55 percent by weight in relation to the participation quantity; as of 1 January 2022, this will increase to 70 percent by weight. In addition, section 16 para. 4 Packaging Act stipulates that at least 50 percent of all waste collected in the light packaging waste collection (usually in a yellow bag or bin) must be recycled. The wording of this target does not refer to a target specific to plastics; nevertheless, the recycling of plastics and plastic composites can contribute to meeting the target.

The targets to which chemical recycling might contribute remain a matter of debate. Chemical recycling should indisputably be subsumed under the concept of recovery within the meaning of section 3 para. 23 Circular Economy Act, as the main outcome of chemical recycling is that the waste is put to a useful purpose within the plant or in the wider economy, either by replacing materials that would otherwise have been used to fulfil a certain function, or by preparing the waste in such a way that it fulfils this function. This should generally be the case for the products generated by chemical recycling (e. g. syngases, oils, monomers).

Provided that packaging waste which falls under the material plastics obtained from system collection is recycled appropriately, the input quantities can contribute to meeting the recovery input target for plastics within the meaning of section 16 para. 2 sentence 2 Packaging Act. The same applies to other waste streams for which recovery targets exist.

For chemical recycling to contribute to the achievement of recycling targets, it would have to process the input waste into products, materials or substances to be used either for their original purpose or for other purposes (section 3 para. 25

Circular Economy Act). Energy recovery and processing into materials intended for use as fuel or for backfilling are not considered to constitute recycling. Insofar as the products of a chemical recycling process are used as a basic material in the chemical industry and are not intended for use as fuel, such a process fulfils the requirement of preparation for a different purpose within the meaning of section 3 para. 25 Circular Economy Act. In concrete terms, chemical recycling products include syngas, oil and monomers from plastic waste, i. e. materials or substances differing from the original material. In this respect, such processes fall under the definition of recycling. It would therefore be possible for chemical recycling to contribute to the recycling target under section 16 para. 4 Packaging Act (50 percent by weight target in proportion to the quantity collected) and, in the case of plastic composites, for meeting the recycling target under section 16 para. 2 sentence 1 no. 6 Packaging Act. The same would also apply to other waste streams for which recycling targets have been set.

The definition of mechanical recovery places extremely high demands on recovery processes. According to section 3 para. 19 Packaging Act, the process of mechanical recovery replaces new material of the same substance or the material remains available for further material use. By explicitly mentioning mechanical recovery, and not recycling, in the target pursuant to section 16 para. 2 sentence 3 Packaging Act, this regulation is intended to ensure that a significantly larger proportion of the waste produced is fed into such high-quality recovery processes. According to the legal interpretation of section 3 para. 19 Packaging Act by the Federal Ministry for the Environment, Nature Conservation and Nuclear Safety (BMU) [13, 14] and the German Environment Agency (UBA), chemical recycling is not recognised as equivalent to mechanical recovery for the known processes, as the material plastic is not preserved, but is converted into other substances (e. g. pyrolysis gas, oil, monomers). These substances are used, for example, as basic materials in the chemical industry, where they replace primary materials. Whether packaging plastics or other products are again produced from these substances in a subsequent process is no longer relevant for the legal assessment of the chemical recycling process. This is because the result of the chemical recycling process (here: pyrolysis gas, oil, monomers) neither replaces new identical material

(here: plastic), nor does the packaging plastic remain available for further material use as such. Rather, the products of chemical recycling replace primary materials (e.g. crude oil), which would otherwise be obtained from fossil raw materials and can be used, for example, as raw material for the production of

many chemical products. It is therefore not possible to include the quantities fed into chemical recycling processes in the 58.5 percent target for mechanical recycling (63 percent as of 2022) in accordance with section 16 para. 2 sentence 3 Packaging Act.

4. Conclusions

The potential advantages offered by all chemical recycling processes include the possibility of removing pollutants from the cycle, the use of plastic fractions that cannot be mechanically recycled, and the wide range of applications for the resulting raw materials within the chemical industry. Feedstock recycling processes could offer a viable alternative for plastic waste that cannot be processed mechanically and that has so far been used for energy recovery. Like mechanical recycling processes, they are considered to be part of (material) recycling and thus take priority over energy recovery in the waste hierarchy. Successful implementation of feedstock recycling could make plastics production more sustainable overall as a supplement to mechanical recycling processes, specifically for waste that cannot be mechanically recycled.

The data currently available suggest that mechanical recycling is generally more advantageous than chemical recycling in both ecological and economic terms, as it involves simpler recovery processes (e.g. fewer additives and less energy used). This is reflected in the design of the recycling targets in the Packaging Act.

Before we can definitively evaluate chemical recycling from an ecological perspective, further time and research are needed to demonstrate the suitability of the techniques involved as well as the ecological advantages of these processes in comparison with energy recovery and mechanical recycling.

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