Fraunhofer CCPE

FRAUNHOFER CLUSTER OF EXCELLENCE CIRCULAR PLASTICS ECONOMY CCPE

POSITION PAPER

RECYCLING TECHNOLOGIES FOR PLASTICS

OBERHAUSEN | SEPTEMBER 2021

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In our position papers, we address topics that are of current concern to society, science and industry. As researchers, we want to take a stand and contribute to objectification in emotional debates. At the same time, we want to show whether and how we can contribute to solving societal challenges. Our position papers are developed jointly by the employees in the Fraunhofer Cluster of Excellence Circular Plastics Economy CCPE - behind a position paper is an opinion-forming process involving several institutes. The present position paper „Recycling Technologies for Plastics" was developed by the Fraunhofer Institutes UMSICHT, ICT and IVV.

Fraunhofer Cluster of Excellence Circular Plastics Economy CCPE

The Fraunhofer Cluster of Excellence Circular Plastics Economy CCPE (Fraunhofer CCPE) combines the expertise of six institutes of the Fraunhofer-Gesellschaft and cooperates closely with partners from industry. Together, we work on systemic, technical and social innovations, focusing on the entire life cycle of plastic products.
Under the leadership of the Fraunhofer Institute for Environmental, Safety and Energy Technology UMSICHT, the following research institutes have joined forces: the Fraunhofer Institute for Applied Polymer Research IAP, the Fraunhofer Institute for Chemical Technology ICT, the Fraunhofer Institute for Material Flow and Logistics IML, the Fraunhofer Institute for Process Engineering and Packaging IVV and the Fraunhofer Institute for Structural Durability and System Reliability LBF.

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Recycling technologies for plastics

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Please cite the position paper as follows:
Hofmann, A.; Franke, M.; Betsch, F.; Rieger, T.; Seiler, E.; Mäurer, A.: Recycling technologies for plastics - position paper, Fraunhofer Cluster of Excellence Circular Plastics Economy CCPE (Ed.), Oberhausen / Sulzbach-Rosenberg, September 2021

State of the document: September 2021

BACKGROUND

Without plastics, many achievements of our modern society would be inconceivable. They make our food last longer (airtight), their low weight as transport packaging for goods and as vehicle components saves fuel and thus CO_2, or in their application as fiber-reinforced composite materials in wind turbine rotors they enable the generation of CO_2-neutral energy.

The production of plastics consumes fossil resources and energy, 4 to 6% of today's oil production is needed for plastics [PlasticsEurope-2017]. This share is expected to increase to 20% by 2050 [Roman-Raminez-2019]. Significant CO_2 emissions are generated along the production chain, and at the end of the product life cycle, the volume and diverse types and compositions of plastics pose a recycling challenge. If disposed of improperly, they also damage ecosystems, e.g. as microplastics in the ocean [UMSICHT-2018].

In 2019, approximately 12.1 million tons of plastics were consumed in Germany, and 6.23 million tons were generated as post-consumer or post-industrial waste. Of these 6.23 million tons, 2.93 million tons (46.6%) were recovered as input in recycling plants. Their rejects and the remaining 3.31 million tons (52.8%) were recovered for energy, i.e., used in waste-to-energy plants or as refuse-derived fuel [Conversio Market-2019].

Compared to the material share of 2.92 million tons, only 0.01 million tons were recycled raw materially (e.g. by chemical recycling) in 2019, which corresponds to a share of 0.2%. This represents the lowest share of feedstock recycling in the last 15 years. In the years 2007 to 2019, between 0.05 million tons and 0.07 million tons were recycled.

During energy recovery, e.g. in the form of incineration, the carbon bound in the plastics is released in the form of heat and CO_2. The average fuel utilization efficiency, i.e. the proportion of the energy content of the waste that can be converted into usable electricity and heat, in the current recycling path of waste incineration plants and RDF power plants is only about 45% (waste incineration plants) to 52% (RDF power plants) on average throughout Germany [Flamme et al.-2018].

In mechanical and chemical recycling, new plastics are produced from used plastics. The carbon thus remains in the utilization cycle. For the recycling of plastics to have a positive climate protection effect, the CO_2 footprint of recycling must not be greater in balance sheet terms than the summed CO_2 footprint from the primary extraction of crude oil and incineration at the end of the product life cycle.

SCOPE OF CONSIDERATION

This position paper addresses material and feedstock (chemical) processing technologies for plastics that are currently under development, in particular those processes that are the research focus of the Fraunhofer Cluster of Excellence Circular Plastic Economy (Fraunhofer CCPE). Mechanical recycling technologies that are already state of the art are not addressed.

Recycling processes are only one solution building block for the transformation from a linear to a circular plastics economy. At Fraunhofer CCPE, approaches are being investigated along the entire life cycle of plastics, including in the areas of design for recycling, development of circular (bio)polymers, research on additives for improving recyclates, plastic substitution and avoidance, transport and logistics, up to circular business models and system considerations [Fraunhofer CCPE-2020].

QUESTIONS AND POSITIONS

OVERVIEW: HOW CAN PLASTICS BE RECYCLED?

A number of methods and processes exist for recycling of plastic waste. They can be divided into two groups:

Mechanical recycling technologies preserve the chemical structure of the plastic [Janz-2020]. These are established processes that are considered state of the art [Ragaert-2017]. Post-consumer or post-industrial plastic waste is thereby processed in mechanical or physical sorting, washing and grinding processes. Remelting (extrusion and melt filtration) is used to produce recyclates from end-of-life plastics. The material properties and the composition of the plastic are largely retained. By adding (compounding) additives, for example stabilizers, pigments, flame retardants or flow improvers, the properties of the recyclate can be adjusted. [Martens-2016]

- State-of-the-art mechanical recycling processes are the best choice for single-grade plastic fractions (thermoplastics).
- With increasing heterogeneity, pollution or contamination of plastic waste, mechanical recycling reaches its limits. Fillers, interfering and harmful substances (e.g. glass, $CaCO_3$, pigments, flame retardants) often cannot be (completely) discharged in sorting, washing and extrusion plants. Certain types of plastics (e.g. thermosets, multilayer plastics, fiber-reinforced plastics) can hardly be recycled.

Unlike mechanical recycling, **chemical recycling processes** change the structure of the polymer chains in the used plastics. They are broken down into smaller molecules or monomers. The corresponding depolymerization processes are mostly solvent-based processes (solvolysis) or thermochemical processes (e.g. pyrolysis). [Lech-2020]

Technically feasible chemical recycling processes are available for nearly all plastics, but only a few have been industrially implemented. Overall economic and ecological evaluations are still pending. These are urgently needed for individual processes as well as process combinations.

- In order to achieve an increase in the recycling of plastics, it is necessary to supplement the mechanical recycling processes with alternative approaches and combinations of mechanical and chemical processes.

WHAT ALTERNATIVE RECYCLING PROCESSES ARE CURRENTLY BEING RESEARCHED?

In the following, processes for solvent-based polymer recovery and depolymerization (chemical recycling) of plastic waste are discussed in more detail.

In the field of advanced mechanical processes, the use of solvents for the separation of plastic waste is being investigated [r+Impuls-2020], [IVV-2020]. In contrast to solvolysis, it is possible to recover complete polymers with these processes. In this respect, they also belong to the mechanical recycling processes. The properties of the recovered polymers are largely retained because the individual plastics are dissolved from each other „as a whole" and interfering and harmful substances are separated in the process [Schlummer-2020], [Schlummer-2018], [Mäurer-2009]. The processes primarily target typical packaging materials made of mixed plastics, including multilayer films made of polyethylene (PE) and polyamide (PA) or polypropylene (PP) and polyethylene terephthalate (PET). The starting polymer can be recovered in almost its original state.

In **chemical recycling**, a distinction is made between solvolysis and thermochemical processes. Solvolysis includes individual processes such as alcoholysis, glycolysis or hydrolysis. Solvolysis is particularly suitable for polyaddition and polycondensation polymers (e.g. PET, PA, PLA, PUR). The feedstock is broken down into monomers, monomer derivatives and oligomers with the addition of a solvent, a depolymerization reagent, a catalyst and heat. These recyclates form starting materials for new polymer systems [Simòn-2018]. The processes are economical feasible for single-grade polycondensation plastics. The monomers produced during solvolysis are suitable for renewed polymer synthesis. Solvolysis reaches its limits when the feedstock is very contaminated. The handling of the solvents is complex [Hohenhorst-2013], [IN4climate-2020].

Thermochemical conversion processes include pyrolysis and gasification processes.

Pyrolysis or catalytic cracking (pyrolysis with the addition of catalysts) takes place at over 300 °C in the absence of oxygen. From mixed plastics, short-chain hydrocarbons are produced as oil, gases (e.g. CO, CH_4 or H_2) and solids (carbon) as products. The oil can be fed directly into petrochemical processing chains or other synthesis processes and processed into new plastics or other chemical products [IN4climate-2020], [Crippa-2019]. So far, gas and solids can only be used energetically, for example to meet the process' own energy demand. The use of the gas phase as feedstock would require further processing steps.

Pyrolysis is very flexible in terms of the feedstock. Recovery of inorganic components such as metals [Gagendta-2020] or fibers contained in carbon fiber reinforced plastics (CFRP) or other composite materials is possible [CFRP Valley-2020]. Post-treatment of pyrolysis products is necessary: the more inhomogeneous the feedstock, the broader the spectrum of chemical components. This leads to additional effort to extract chemicals in sufficient purity [IN4climate-2020] [UMSICHT-2020].

In contrast to pyrolysis, **gasification** is carried out with the addition of e.g. oxygen or steam at temperatures between 700 and 1600 °C and a pressure between 10 and 90 bar. In the gasification reaction, the plastics are broken down into basic chemical building blocks such as CO, H_2 and also CH_4. These basic chemical building blocks can be fed into petrochemical processing chains but also into other synthesis processes. Gasification processes of plastic waste have high energetic and process engineering requirements that can only be realized in large-scale plants. This, in turn, requires large feedstock volumes at a single site, combined with high transportation costs and adverse environmental impacts. Purification of the product gas is particularly important, since the downstream processes are predominantly catalytic and therefore very sensitive to impurities.

WHAT IS THE STATE OF DEVELOPMENT OF CHEMICAL RECYCLING TECHNOLOGIES?

Driven by an increasing demand in polymer chemistry for recycling-based basic materials, there are numerous development projects in the field of chemical recycling on the part of both science and industry. They range from laboratory and pilot plants to individual commercial projects. There is only incomplete information with regard to maturity and efficiency levels, plant sizes and economic viability.

Our own market analysis (see Fig. 1) shows that it is mainly large companies from the polymer production as well as the chemical industry that are making strategic investments in the field of chemical recycling. These include corporations such as SABIC, BASF, Indorama, Eastman Chemicals, Covestro or LyondellBasell [ClosedLoopPartners-2020].

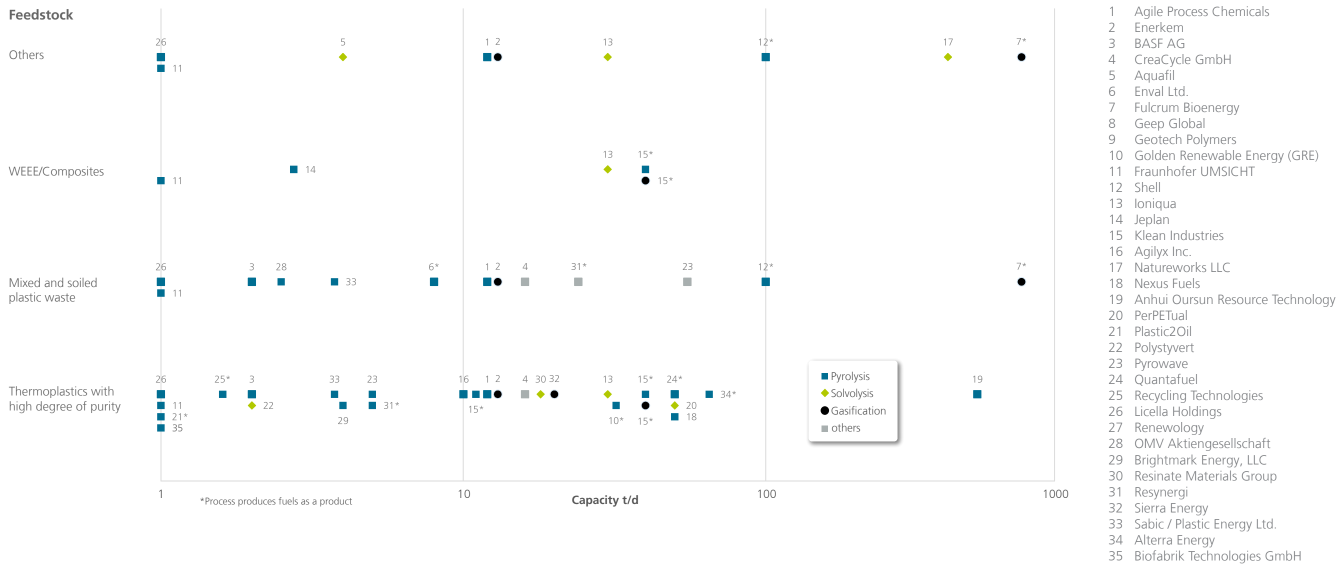

Fig. 1: Overview of current initiatives for the demonstration and commercialization of chemical recycling processes. Shown are projects from a TRL 7 with feedstocks and capacity (partly estimated).
Source: Own research, ClosedLoopPartners-2020, IN4climate-2020.

In particular, pyrolysis, catalytic cracking and chemical depolymerization (solvolysis) are the focus of current technology developments. The pyrolysis and catalytic cracking technologies are the most advanced in terms of the range of feedstocks. Chemical depolymerization is currently available for PET or other addition or condensation polymers with a high Technology Readiness Level (TRL) [Lech-2020].

It is to be expected that the share of processes available on the market and thus the share of chemically recycled plastic waste will increase in the coming years. Despite specific strengths, none of the processes alone is suitable for recycling of the wide range of (heterogeneous) plastic waste and returning high-quality recyclates, monomers or basic chemicals to the cycle. It is necessary to select the most effective process or process combination for the different input streams.

WHAT CAN ADVANCED RECYCLING PROCESSES ACHIEVE OVERALL?

Chemical and advanced mechanical processes allow **high-quality recycling** of plastic waste. This makes it possible to **improve the quality** of recyclates and even to produce polymers in virgin material quality. This is accompanied by a wider range of applications, also in the area of higher-value products (e.g. high-quality surfaces, medical products, toys, food packaging, etc.) and potentially higher recycling rates.

Chemical recycling processes are capable to recycle a **wide range** of plastics and plastic mixtures, including polyolefins, polystyrene, PET, PA, PC, ABS plastics, various duromers (e.g. phenolic resins, epoxy resins), mixed and/or contaminated plastic waste such as municipal waste or multilayer packaging, shredder light fractions from the automotive industry, used tires, WEEE (waste electrical and electronic equipment), waste from the construction sector, composites such as glass- or carbon fiber-reinforced plastics, or even bio-based plastics such as PLA.

If the process is appropriately controlled, depolymerization can in principle lead to the **removal of pollutants and impurities**, e.g. halogenated, persistent organic flame retardants such as brominated diphenyl ethers (deca-DBE), which occur in end-of-life vehicle waste and WEEE, can be separated [Hense-2015]. Corresponding processes for the purification of products from chemical recycling still have to be optimized and transferred to stable continuous operation.

Overall, advanced recycling technologies can help to avoid the disposal on landfills or energetic utilization of plastics that could not previously be recycled. In the future, technical developments can further reduce the cost of pretreatment (sorting, washing). In addition, recycling plastics can reduce dependence on crude oil imports and waste exports.

WHAT FRAMEWORK CONDITIONS ARE REQUIRED FOR CHEMICAL RECYCLING PROCESSES?

Acceptance and market penetration of chemical recycling are mainly dependent on regulatory, economic and ecological factors, but also on further technical development and proof of large-scale continuous operation.

In Europe, the recycling of plastics is **heavily regulated by law**. Both the EU and Germany in particular are constantly increasing their quotas.[1] The EU Waste Framework Directive and the corresponding German Closed Substance Cycle Waste Management Act as well as the EU Packaging Directive give recycling by mechanical and chemical processes priority over energy recovery. In contrast, the German Packaging Act (VerpackG) defines a recycling quota (sum of material and energy recovery) for plastic packaging subject to system participation and a separate quota for material recovery only. The latter amounts to at least 58.5% (currently) or 63% (from 2022 on). According to the legal opinion of the German Federal Ministry of the Environment, Nature Conservation and Nuclear Safety, the definition of mechanical recycling in the Packaging Act excludes chemical recycling, which is thus equated with energy recovery in terms of the quota. In addition, there has been a general lack of a legal definition of the term "chemical recycling" to date.

- Since chemical recycling processes are also capable of providing secondary raw materials for plastics production, the current mechanical recycling quota in the German Packaging Act should be replaced by a recycling quota that is open to all technologies. This would promote technical innovations in the recycling of packaging and allow advanced processes to enter the market.

The economic viability of chemical recycling processes cannot be conclusively assessed at present. In addition to the technical effort involved, this depends heavily on the type of used plastic, the type of new product and other ecological, economic and regulatory constraints. A low oil price, for example, inhibits efforts to substitute fossil raw materials with secondary raw materials. In the past, this has led to recycling plants becoming uneconomical [Material Economics-2019]. With the planned introduction of recyclate quotas for various application areas of plastics, the demand for plastic recyclates could be decoupled from the oil price and thus stabilized.

The basic technical feasibility of chemical recycling processes has been demonstrated. The chemical recycling plants in operation are mostly in pre-commercial pilot or demonstration scale. Reliable facts on investment and operating costs, product quantities and qualities, their sales structures and prices that can be realized are not available to date.

[1] The EU Circular Economy Package has initiated extensive changes to European waste legislation, which also affect the area of plastic waste. The Waste Framework Directive (ARRL) provides for increasing recycling rates for municipal waste (55% by 2025, 60% by 2030 and 65% by 2035). The recycling rate target for plastic waste has been significantly increased in the European Packaging and Packaging Waste Directive (94/62/EC) (50% by 2025 and 55% by 2030). In addition, Germany is implementing the targets even more ambitiously with the Packaging Act (VerpackG) and has required a recycling rate for plastic packaging of 58.5% since 2019 and 63% as early as 2022.

Decisive for the economic viability of recycling processes are, among other things, the quantity of feedstocks available. The transformation of German plastics production from around 18 million t/a [PlasticsEurope-2017] alone to recycled basic chemicals originating from secondary raw material sources would require a redirection of waste streams that have so far been used for energy recovery. For chemical recycling to be a useful complement to mechanical recycling, an appropriate material flow management must be established, e.g. to feed residues from mechanical recycling to chemical processes (process cascades).

The demand side for plastics from chemical recycling can be classified as good. The chemical industry, consumer goods manufacturers and the automotive industry are all interested in using chemically recycled products, provided that the quality of virgin materials is achieved [Endres-2020].

- Reservations about plastic recyclate are often due to the lack of reliable and comprehensive quality standards. Existing and new standards should be adapted and expanded with regard to chemical recycling processes.

The aspect of **environmental sustainability** of chemical recycling processes is addressed below.

CAN CHEMICAL RECYCLING MAKE A CONTRIBUTION TO ENVIRONMENTAL AND CLIMATE PROTECTION?

In principle, chemical recycling processes allow the **targeted removal of pollutants** from the material to be recycled, but there is still a need for research in this area for industrial application. Processing residues containing pollutants must be disposed of in a controlled manner. If additional chemicals (e.g. solvents) are used in the process, these must be recycled or disposed of properly.

The plastics used in Germany cause **GHG emissions** of approx. 30 million tons CO_2-equivalent per year from their production and 9 million tons of CO_2 from the incineration of plastic waste (for comparison, a total of 810 million tons GHG were released in Germany in 2019) [Umweltbundesamt-2021]. With mechanical or chemical recycling, plastic is kept in the cycle. Incineration and landfilling are avoided. On the one hand, this reduces the amount of new petroleum-based plastics (**substitution effect**), and on the other hand, the carbon remains bound in the product (**carbon sink**) and does not enter the environment as CO_2.

It should be noted that conventional mechanical recycling inevitably leads to a degradation of the material properties of the plastics (downcycling). In addition, the quality of the recyclate deteriorates with each further cycle [Martens-2016]. This leads to the fact that virgin plastics cannot always be replaced 1:1 without additional treatment. The function of the carbon sink is thus only maintained (at least for the period of use) if a durable plastic product is created from the recyclate.

A frequently asked question in the field of chemical, especially thermochemical recycling processes such as pyrolysis or gasification, is that of **energy and life cycle assessment**.

It should be noted that a comparative overall energy or life cycle assessment of chemical and material processes in continous operation does not yet exist. Industry-specific scenario analyses (e.g. for the chemical sector) estimate the CO_2-savings potential as a result of integrating chemical recycling processes positively, but always emphasize the dependence on the emission factor of the underlying energy mix [Bazzanella-2017] [Accenture-2017] [Material Economics-2019]. Model calculations also see an advantage of chemical recycling processes compared to incineration with regard to climate effects, if it is possible to recycle plastics that have been incinerated up to now [Meys-2020].

- An overall ecological, comparative analysis of specific recycling processes or process combinations for specific used plastics still has to be carried out.
- A partial substitution of petroleum-based basic chemicals for the plastics industry by chemical recyclates, e.g. based on plastic waste, appears technologically possible.

RESEARCH AGENDA RECYCLING TECHNOLOGIES

Innovative recycling technologies can help improve the plastics circular economy. In particular, chemical processes could be a complementary building block for higher-value plastics recycling. But there is still need for research and development. The following outlines what Fraunhofer CCPE believes is a reasonable research agenda for enhanced recycling processes:

1. Analysis of waste containing plastics

For heterogeneous plastic waste, it is necessary to collect more data on its composition, especially with regard to the polymers, additives and contaminants it contains. On this basis, the best possible treatment process can be selected. Above all, faster, more accurate and automated sensor technology and analytics are the scientific focus here. In addition, the digitization of waste management value chains should be advanced.

2. Transparency about economic and ecological effects through long-term operation

A reliable evaluation of recycling processes requires the long-term operation of corresponding plants on a demonstration scale. The construction and operation of corresponding demonstrators should be promoted. These can then provide meaningful and comparable data for mass, energy and carbon balances as well as technical, economic and ecological assessments.

3. Dynamic evaluation models for waste treatment

Building on the primary data and by applying specific criteria, it is possible to evaluate which recycling technology is best suited for which waste-streams under which general conditions, e.g. based on material flow size, heterogeneity, contaminations, local factors, economic efficiency, life cycle assessment etc. A dynamic evaluation model then allows adaptation to different target scenarios (e.g. „highest possible product quality“, „minimize energetic utilization“, „minimum CO_2-emissions“ etc.) and changing boundary conditions (e.g. recyclate demand, energy costs, volume availability).

4. Combination of recycling technologies

One recycling technology alone is not able to sustainably manage all plastic waste. Based on data and evaluation models (steps 1-3), it therefore makes sense to combine several recycling technologies into integrated systems. In this way, the best possible process combination is available for the respective single-origin-feedstock or mixed waste.

5. Automated, AI-based recycling processes

With the help of semi-autonomous, AI-based systems, the type of pretreatment is controlled and a decision is made as to which individual fractions are recycled via mechanical, solvent-based or chemical recycling. Rejects from each process are added to the next most robust process. With automated process cascades, maximum energy and raw material efficiency can thus be achieved and the optimum process combination identified.

6. Optimization of products from recycling

In both mechanical and chemical recycling, research and development are necessary to improve the qualities of the recycled products. On the one hand, (new) recyclate standards and quality controls are decisive here; on the other hand, exchange with industry is necessary in order to achieve certain, application-specific material properties. There is also a need for research in the area of processing intermediate products (e.g. pyrolysis oils) for further processing up to virgin plastics.

At Fraunhofer CCPE, the competencies of various Fraunhofer institutes are combined in a joint research program, from polymer development, recycling technologies, the evaluation of these recycling technologies, the reuse of recyclates to the development of customized business models.

Fraunhofer Competence Landscape

Recycling technologies

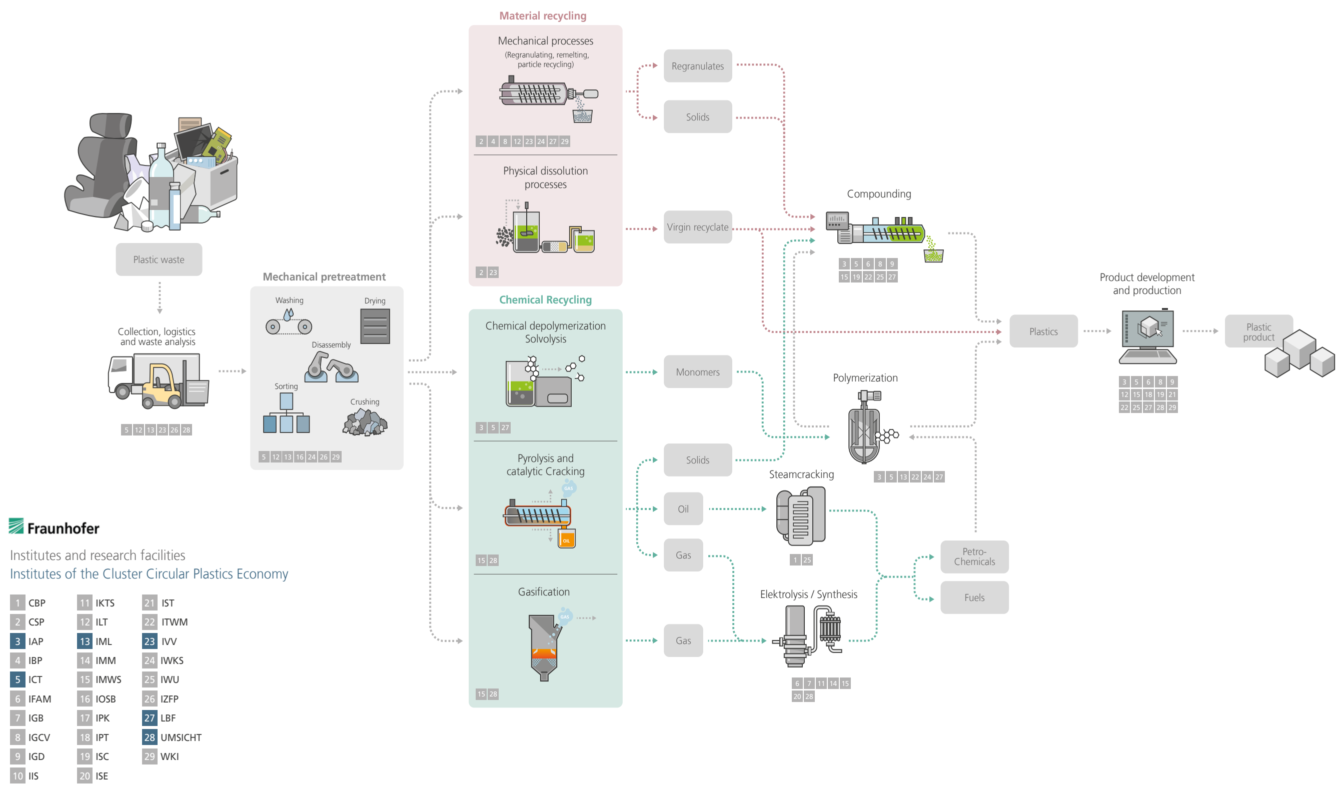

Fig. 2: Fraunhofer competence landscape recycling technologies

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