

Chemical Recycling for Plastic Waste: Status and Perspectives[‡]

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Abstract

Industrial-scale mechanical recycling of plastics has been established for years, but has technical and economic limits. Chemical recycling processes lead back to monomers or to the raw materials, so that in the end new goods can be produced for all areas of application of plastics. The variety of chemical recycling processes is large. The capacities of the plants are still low today. The profitability of the plants is strongly influenced by the price of oil; the profitability limit is currently between 50 and 60 US \$ per barrel.

Keywords: Chemical recycling, Oxyfuel combustion, Plastic waste recycling, Pyrolysis, Solvolysis

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1 Introduction

Plastics have become an integral part of everyday life. Consumption in Germany in 2019 was 12.1 million tons [1]. Polyethylene, polypropylene, and polystyrene account for more than 50 % of this amount. Staudinger first reported on polymerization some 100 years ago [2]. For this he was awarded the Nobel Prize in Chemistry in 1953. In 1950, 2 million tons of plastics were produced worldwide, in 2015, 380 million tons. Geyer et al. have calculated that from 1950 to 2015 around 8300 million tons of plastics were produced (primary production) [3]. Secondary material hardly plays a role in this analysis, because just 9 % of it was recycled. Most of this (79 %) ended up in landfill or in the environment (e.g., 150 million tons in the oceans [4]), and 12 % was incinerated.

In the EU, the demand for plastics in 2017 was 51.2 million tons. Almost 40% of it was used as packaging material [5], which is a rather short-lived application compared with plastics in the construction sector (20%) or in the automotive industry (10%) and is therefore due for disposal after a short period of use. The recycling pathways for plastic packaging waste vary greatly in the individual EU member states; on average, the EU states achieved recycling of almost 80% in 2017. In countries with recycling rates over 80%, thermal recycling usually dominates. In Germany, the shares of mechanical recycling and thermal recycling are about the same.

The limitations of mechanical recycling result from the deterioration of mechanical properties due to oxidative degradation reactions and inconsistent product quality [6]. In addition, there are high costs and great effort when mixed plastics and composites have to be separated. Chemical recycling has not yet played a major role. For 2018, PlasticsEurope reported 10 600 tons in Germany and 3600 tons in Italy for chemical recycling (0.2 and 0.1%, respectively) [7]. This proportion will increase in future, as a look at current literature on the subject shows [8–12].

This review focuses on pyrolysis and solvolysis processes. The procedures and their most important features are described in tabular form. In conclusion, benefits and areas of concern and need for research are discussed.

2 Overview of Chemical Recycling Processes

Recycling is a waste treatment measure that ranks third in the 5-level waste hierarchy according to the European Waste Framework Directive. Waste avoidance and preparation for reuse are located above this. Reuse of plastic products after

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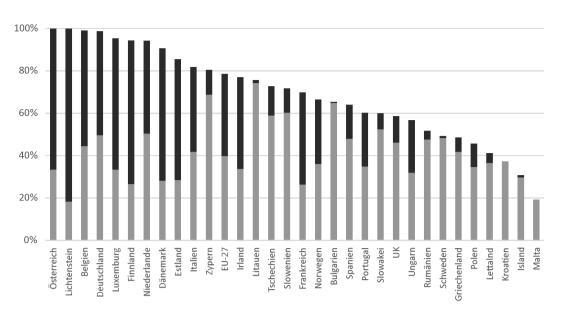
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processing is conceivable, but is the exception rather than the rule. The reuse of production waste (in the simplest case, cuttings) in the manufacture of plastic products is referred to as primary recycling [14] and is the best method for recycling in all respects. The products from mechanical processing methods are used as raw materials for new plastic products. However, mechanical recycling processes (secondary recycling) have their limitations with highly mixed and contaminated plastic waste. Here, chemical recycling processes can help to close the gaps in the cycle. Combustion (particularly in the case of oxygen enrichment, oxyfuel) can also make a contribution, as it is easier to separate CO_2 here [15]. However, incineration counts as "other recycling" in the waste hierarchy and takes the fourth place before disposal.

Chemical recycling of plastics is generally understood as an industrial process based on the breakdown (decomposition) of long-chain organic macromolecules into short-chain organic molecules, monomers, or their components with the subsequent use of at least part of these breakdown products in manufacturing new plastics. Depending on the chemical principle or process implemented for the decomposition, a distinction is made between hydrolysis, solvolysis, hydrocracking, catalytic cracking, pyrolysis, and gasification. Although incineration is also a chemical use process, this process cannot really be considered recycling if the resulting combustion products (essentially CO_2 and H_2O) are not recycled, which they are not in the majority of incinerators.

The use of chemical recycling products to produce fuels (petrol, diesel) is possible and widespread in some regions (e.g., India) and can also be considered a kind of recycling. Chemical recycling is technically much more demanding than mechanical recycling. However, it offers flexibility in the production of the new plastic and even allows the production of plastics from recycled raw materials in the quality of new goods ("virgin quality"), which is definitely not the case for the mechanical recycling. In the following sections, pyrolysis and solvolysis are considered and analyzed in detail. Fig. 2 shows how reuse and different recycling processes fit into the whole life cycle of plastics.

2.1 Pyrolysis

During pyrolysis, long-chain polymer molecules are split into short-chain components by supplying thermal energy in the absence of oxygen (inert atmosphere or vacuum). As a result of pyrolysis, a mixture of gas (e.g., H_2 , CH_4 , C_2H_6 , CO), liquid (e.g., C4–C10 compounds), and solid-phase products (e.g., paraffins, wax, carbon) is formed [12, 16]. The exact composition of the pyrolysis products depends both on the raw material (plastic mixture) and on the pyrolysis conditions (temperature, catalyst, carrier gas, pressure). Liquid pyrolysis products are often referred to as pyrolysis oil and can either be added to the crude oil stream in cracking plants or, after fractionation, used as a raw material in chemical production (plastics production, surfactants, plasticizers, etc.).

In some process variants, hydrogen is required to control the output [9, 17]. Gas-phase products are used in most plants as energy carriers for pyrolysis, but concepts are known in which gas-phase products are the main part of the process yield. More lucrative exceptions are poly(methylmethacrylate (PMMA) and polystyrene (PS), which can be pyrolyzed into the monomers MMA and styrene, respectively. An overview of the current pyrolysis plants and projects is given in Tab. 1.

This overview clearly demonstrates that pyrolysis is already used for recycling or as a source of raw materials in various countries. It is a fairly well-known and quite widespread technology. The overall pyrolysis capacity was approximately

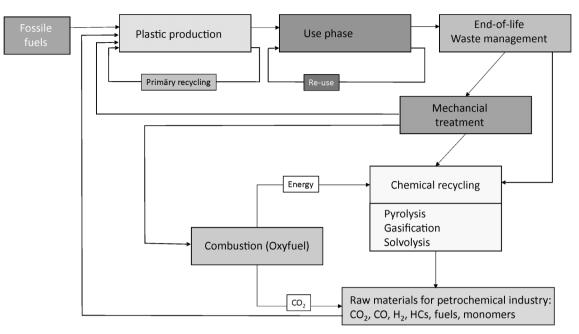


Figure 2. Chemical recycling as part of a circular economy for plastic materials.

Company	Country	Process	Raw material	Process temperature	Products	Capacity [tpa]	Partner	Status/summary
Sulapac	FIN	Catalytic pyrolysis	Biocomposite	400-600 °C	Liquid HCs		VTT	The producer of biodegradable products is developing a pyroly- sis plant in cooperation with VTT.
Indaver	В	Pyrolysis	Unsorted plastic waste, PE, PP		Liquid HCs, waxes, paraf- fins	7000	Univ. Ghent, Univ. Ant- werp, INEOS	The waste disposal company operates a pilot plant with a capacity of 7000 tpa; an industri- al plant with 15 000 tpa is in planning. A calculation for 120 000 tpa was executed [18].
PowerHouse Energy	UK	Pyrolysis	Unsorted plastic waste	1000–1200 °C	CO, H ₂	9000		A demonstration plant was built based on the DMG [®] technology (distributed modular generation) from Pyromex. An additional plant with a capacity of 12 000 tpa is planned.
Recycling Technologies	UK	Pyrolysis	Plastic waste	400–600 °C	Liquid HCs	7000	Total, Nestle, Mars	The company's own technology for the production of smaller or mobile plants (suitable for container).
Plastic Energy	UK	Pyrolysis	PE, PP, PS	400 °C	Liquid HCs	7000	SABIC	Modular technology is applied in Spain at sites in Seville and Almeria.
Itero (formerly CGC)	UK	Pyrolysis	Plastic waste	350–500 °C	Liquid and gaseous HCs	27 000		Itero operates a pilot plant with a capacity of 5500 tpa. An industrial pyrolysis plant in the Netherlands is slated to start up in 2022.

Table 1. Overview of the developers of pyrolysis plants, projects, and	operators.
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Company	Country	Process	Raw material	Process temperature	Products	Capacity [tpa]	Partner	Status/summary
Susteen Technologies	DE	Thermo- catalytic reforming (TCR [®])	Plastic waste, organic waste	400–500 °C	Liquid and gaseous HCs, coke	4000	Fraunhofer UMSICHT	Demonstration plant using a technology developed by Fraunhofer UMSICHT (Germany). Pyrolysis oil is cata- lytically reformed in a separate step [19–21].
Pyrum	DE	Pyrolysis	Plastic waste, tyres	300–1200 °C	Liquid and gaseous HCs	7000	BASF	The developer of the technology operates a demonstration plant und a pyrolysis plant for waste tyres with a capacity of 10 000 tpa. BASF has announced a 16 Mn EUR investment for two additional pyrolysis plants from Pyrum. BASF uses the pyrolysis oil in the framework of the ChemCycle TM project.
Clariter	PL/LUX	Pyrolysis	Plastic waste	400 °C	Liquid HCs, waxes, paraffins			Pyrolysis products are used as high-value raw materials to produce solvents and oils.
Pohjanmaan Hyötyjätekul- jetus (PHJK)	FIN	Pyrolysis	Unsorted plastic waste		Liquid HCs	4000		The waste disposal company operates smaller plants.
ReOil / OMV	А	Pyrolysis	Plastic waste, PE	450 °C	Liquid HCs	900	Borealis	Developed by ReOil (belongs to the refinery operator OMV), which operates a small demon- stration plant for the pyrolysis of LDPE and heavy oil residues [22].
Splainex	NL	Pyrolysis	Plastic waste, wood, WEEE	500 °C	Liquid and gaseous HCs	35 000		The plant engineering company develops the pyrolysis process and plant concept. The plants are sold globally [23].
Smuda/ Agrob Eko	PL	Catalytic pyrolysis	Organic waste, plastic waste	300-450 °C	Liquid HCs	10 000		The technology was developed by Henryk Smuda; the plant is located in Zabrze, Poland [24].
Leitner Technologies	SLO	Pyrolysis	Plastic waste	400 °C	Liquid HCs	360		Plant engineering company for smaller plants.
Quantafuel	N/DK	Catalytic pyrolysis	Plastic waste	420 °C	Liquid and gaseous HCs, coke	20 000	BASF	The developer of the technology started operating its own plant in Aarhus in 2019. A coopera- tion with Remondis and BASF was announced in 2021. Quan- tafuel will deliver the technology for a 250 000 tpa plant at BASF location Ludwigshafen and will operate the plant starting in 2025. Remondis will deliver the plastic waste; the pyrolysis oil will partly cover the demand of BASF.



Company	Country	Process	Raw material	Process temperature	Products	Capacity [tpa]	Partner	Status/summary
Fuenix Ecogy	NL	Pyrolysis and plasma treatment	Plastic waste, PE, PP	500–800 °C	Liquid and gaseous HCs		DOW	Ecogy [®] plasma treatment delivers the main item of the pyrolysis technology, implemented in the plant located in Weert. In 2019, a cooperation with DOW was concluded with the objective to deliver 100 000 tpa of secondary raw materials, starting in 2025.
Neste	FIN	Pyrolysis	Organic waste, plastic waste	400–600 °C	Liquid and gaseous HCs		Alterra	The refinery operator and pro- ducer of raw materials for the chemical industry is developing a modular pyrolysis technology for plastic waste and renewable raw materials. A demonstration plant was built with the objective to recycle 100 000 tpa of plastic waste starting in 2030.
Cassandra Oil	SWE	Catalytic pyrolysis, mechani- cally acti- vated	Plastic waste, tyres (no PVC)	600 °C	Liquid HCs	15 000	Sacyr	Technology developer for waste disposal and recycling compa- nies with the largest pyrolysis plant in operation (Jerez, Spain, capacity 15 000 tpa). According to Cassandra Oil, the break-even oil price is 25 USD/bbl.
New Energy	HU	Pyrolysis	Tyres	800–1000 °C	Liquid HCs	9000	BASF	New Energy signed a contract with BASF for the delivery of 4000 tpa after the successful pilot phase in Ludwigshafen.
MOL	HU	Pyrolysis, cracking	Plastic waste, organic waste	300–360 °C	Liquid HCs			The refinery operator developed its own pyrolysis process on a pilot scale [25, 26].
Lyondell- Basell	Ι	Pyrolysis	Unsorted plastic waste	< 550 °C	Liquid and gaseous HCs, coke	80	KIT	Operation of the pilot plant MoReTec started in 2020 in Ferrara, Italy. The technology was developed at KIT in Germany [27].
Paterson Energy	IND	Catalytic pyrolysis	Plastic waste	500 °C	Liquid HCs	2000		Paterson Energy operates several plants in India. The process yield is 50 %, which is too low for the current state of develop- ment.
Agile Process Chemicals LLP (APChemi)		Pyrolysis	Plastic waste, tyres		Liquid HCs, waxes, paraffins	10 000	Shell	The plant engineering company developed a pyrolysis process and plant concept. The plants are sold globally.
Rudra Envi- ronmental Solutions	IND	Catalytic pyrolysis	Plastic waste	380-430 °C	Liquid and gaseous HCs, coke	1800		Technology developer for waste disposal and recycling companies [28]
Polycycl Private Limited	IND	Pyrolysis	Plastic waste		Liquid HCs		Ramky	The technology company devel- oped a continuous process for waste disposal and recycling companies and operates a demonstration plant [29].

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Company	Country	Process	Raw material	Process temperature	Products	Capacity [tpa]	Partner	Status/summary
Toshiba Plant System	JPN	Pyrolysis	PS	700 °C	Styrene	1000		Process and plant for waste prevention in the company's own factory.
Toyo Styrene	JPN	Pyrolysis	PS	450–550 °C	Styrene	3000	Agilyx	Technology licenced by Agilyx (USA).
JSW	JPN	Pyrolysis	РММА	400 °C	MMA	n.a.	Arkema	Technology supplier in the research project MMAtwo initi- ated by Arkema (Horizon 2020, budget 6.6 Mn. EUR). Pilot experiments were carried out in June 2020 [30].
Sapporo Plastics / Klean Industries	JPN/ USA	Pyrolysis	PE, PP, PS	400 °C	Liquid HCs	15 000	Toshiba	The technology developed by Toshiba was transferred in 2011 to Klean Industries, which is proceeding to market the technology.
Hitachi Zosen Corporation	JPN	Pyrolysis	PE, PP, PS, Abs, PVC	250–350 °C	Liquid HCs	n.a.		The plant engineering company developed a pyrolysis process and plant concept.
Chiyoda Corporation	JPN	Pyrolysis	PE, PP, PS, ABS, PVC	2-step pyrol- ysis with distillation columns, 340–390 °C	Liquid HCs	6000	Rexol, Fuji	The plant engineering company developed its own pyrolysis process, which was realized in a plant [31]. Rexol is responsible for marketing the pyrolysis products.
Nissan / Mitsui Chemicals	JPN	Pyrolysis	PP, PE, PET	315 °C	Liquid HCs	Pilot phase		Cooperation for the use and prevention of waste in the automotive industry.
Showa Denko	JPN	Pyrolysis/ gasifica- tion	PE, PP, PS	2-step process, 600–700 °C, 1300–1500 °C	CO, H ₂	64 000	Ebara Corpo- ration, Ube Industries	Ebara Ube Process (EUP) delivers H ₂ for NH ₃ production. Showa Denko now uses plastic waste from McDonald's in Kawasaki for the proof of concept.
Nippon Steel	JPN	Pyrolysis in a blast furnace	PE, PP, PS	1200 °C	Liquid and gaseous HCs, coke	200 000		Known technology from the steel industry is applied in Japan for recycling and resource recovery.
Jinan Niutech Environment Technology Corporation	CN	Pyrolysis	Plastic waste	500 °C	Liquid HCs	10 000		The plant engineering company developed a continuous pyroly- sis process and plant concepts for different raw materials (waste plastic, tyres, biomass). The plants are sold globally.
Xinxiang Huayin Renewable Energy Equipment Co., Ltd.	CN	Pyrolysis	Plastic waste	400–450 °C	Liquid and gaseous HCs, coke	3500		The plant engineering company developed a pyrolysis process and plant concept; the plants are sold globally.
Bioland	CN	Pyrolysis	Plastic waste		Liquid HCs			The waste disposal company developed its own pyrolysis process for plastic waste.

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Table 1.	Continued.
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Company	Country	Process	Raw material	Process temperature	Products	Capacity [tpa]	Partner	Status/summary
Agilyx	USA	Pyrolysis	PS, PMMA	450–550 °C	Styrene, MMA	7000	AmSty, Lucite	In their joint venture, Regenyx, Agylix, and AmSty demon- strated an example of an indus- trial cycle. Waste polystyrene is depolymerized to styrene and used to manufacture new prod- ucts. In cooperation with Lucite, research is conducted for a process for a PMMA cycle.
GEN2WTE	USA	Pyrolysis	Unsorted plastic waste (no PVC)		Liquid HCs	5800		Technology was licenced by Gen Tech [41].
Braven	USA	Pyrolysis	Unsorted plastic waste		Liquid HCs	65 000		Braven announced its plans to build a large plant in the USA in 2021.
Green EnviroTech	USA	Pyrolysis	Tyres, unsorted plastic waste		Liquid HCs	35 000		
Nexus Fuels	USA	Pyrolysis	HDPE, LDPE, PP, PS		Liquid HCs, waxes, paraffins	18 000	Shell	Nexus operates its own recycling plants.
Plastic Ad- vanced Recycling Corporation (P.A.R.C)	USA	Pyrolysis	Plastic waste, tyres, waste oil		Liquid and gaseous HCs, coke			The plant engineering company developed a pyrolysis process and plant concept.
Plastic2Oil	USA	Catalytic pyrolysis	Unsorted plastic waste	350-450 °C	Liquid HCs	15 000		Conversion of plastic waste to fuel.
QCI	USA	Pyrolysis	Unsorted plastic waste		Liquid and gaseous HCs, coke	18 000		QCI processes plastic waste and waste tyres to fuel, solvents, and carbon black. An extension of the plant capacity to 100 000 tpa was announced for 2021.
Resynergi	USA	Micro- wave-in- duced pyrolysis	PE, PP, PS	650–700 °C	Liquid and gaseous HCs, coke	3500	University of Minnesota	This start-up company imple- ments microwave-induced py- rolysis plants in 20-foot contain- ers. According to the producer, the cost of a 20-foot container plant is 350 000 USD; a 40-foot container plant costs 2 Mn USD.
RES Polyflow (BrightMark)	USA	Pyrolysis	Unsorted plastic waste		Liquid HCs, waxes, Paraffins	100 000	ВР	A large plant will be built after operation of the first pilot plant. In this project, BP acts as a customer.
Renewology	USA	Pyrolysis	Unsorted plastic waste	400–550 °C	Liquid HCs	3500		The capacity refers to a demon- stration plant in Salt Lake City; a larger plant will be built in Phoenix.
Vadxx/ Alterra Energy	USA	Pyrolysis	Unsorted plastic waste	300-600 °C	Diesel, naph- tha, syngas	25 000	Neste	A continuous pyrolysis process was developed by Vadxx, which is now part of von Alterra Energy.





Company	Country	Process	Raw material	Process temperature	Products	Capacity [tpa]	Partner	Status/summary
Green Mantra	CAN	Thermo- catalytic depolyme- rization	PE, PP, PS	300–600 °C	Waxes, polymers, additives	1100	Sun Chemi- cal, INEOS	Pyrolysis products are used as high-value additives. Additional polystyrene from waste is completely recycled.
New Hope Energy	USA	Pyrolysis	PE, PP, PS		Liquid HCs	340 000	Chevron Phillips	New Hope Energy operates its own pyrolysis plant in Trinity Oaks Tyler. The plant capacity will be extended. Chevron Phil- lips acts in the cooperation as customer. Circular polyethylene is already produced from the pyrolysis oil Marlex [®] Anew TM ; the volume of production will reach 450 000 tpa in 2020.
Adherent Technologies	USA	Pyrolysis, solvolyse	PE, PP, PS, epoxy	300–600 °C	Liquid and gaseous HCs			A pilot plant was built based on the technology developed by Adherent. The focus was placed on recycling polymer fibers by Adherent Technology [32].
Pyrowave	CAN	Micro- wave-in- duced catalytic pyrolysis	PS	300–400 °C	Styrene		INEOS	The technology developer delivers microwave reactors for pyrolysis plants.
Encina Technologies	USA	Pyrolysis	РР	300–400 °C	Liquid HCs	160 000	Braskem	The technology developer plans to build a pyrolysis plant in 2023 to produce recycled PP from Braskem.
BP	DE	Pyrolysis	Unsorted plastic waste		Liquid HCs		SABIC	BP and Sabic signed an agree- ment with the objective to coop- erate in the field of the circular economy. BP will deliver ethyl- ene produced from pyrolysis oil to SABIC as raw material for products from the TRUCIRCLE [™] portfolio.

1 million tons in 2021. According to numerous announcements (Chevron Philips, Neste, BP, Quantafuel/BASF), this capacity will increase by at least a factor of 4 by 2030. Though this is just a small share of the worldwide manufactured 370 million tons of plastics, the growth rate underlines the increasing importance of pyrolysis for plastics recycling. Most of the companies mentioned above (see Tab. 1) already operate pyrolysis plants with listed capacities. These plants can be classified by raw material mix, product quality, capacity, and operating company as follows:

Raw material:

1. Pure plastic material (polyethylene (PE), polypropylene (PP), polystyrene) or defined mixtures of plastic materials (PE and PP): mainly comparatively old plants or plant concepts without the possibility to handle polyvinyl chloride (PVC). A more or less defined raw material allows com-

paratively easy operation of pyrolysis units and leads to a comparatively stable product.

- 2. PS and poly(methylmethacrylate) (PMMA) depolymerization plants (Toshiba Plant System, Toyo, JSW, Agilyx): this is a "real" chemical recycling of polymers down to monomers, which allows any further use of monomers and fulfils the concept of circularity.
- 3. Mixture of plastic materials or mixture of plastic materials and cellulose: these modern plants are flexible regarding the raw material, due to sorting before pyrolysis and/or due to the dechlorination of pyrolysis products. However, the operators of such plants must either find customers for products with very broad specification or implement measures to increase product stability/homogeneity. Product quality:

- 1. Pure monomers (styrene, methyl methacrylate (MMA)): see above
- 2. H_2/CO mixture (PowerHouse Energy, Showa Denko): in these plants, pyrolysis (gasification in case of Showa Denko) is carried out at quite high temperatures (800 °C and more) to optimize the yield of H_2/CO . The choice of these products is because their market prices are higher than for hydrocarbons. However, high pyrolysis temperature makes these processes challenging and energy-intensive.
- 3. Mixture of gas-, liquid-, and solid-phase products: this is the case for most pyrolysis plants. Some operators try to increase the share of liquid-phase products. Under optimized process conditions, the yield may reach 90 %.
- Plant capacity:
- 1. Up to 10 000 tpa: these are usually demonstration, proofof-concept, or research plants. They may be used commercially, but the main purpose of these plants is to attract investors or partners for larger projects. Another application for plants of this capacity in some regions (India, Finland) is manufacturing diesel fuel.
- 2. 10 000–30 000 tpa: this is usually the size range for commercial plants for converting local plastics waste into liquid hydrocarbons for further use of these hydrocarbons by local industry; the majority of pyrolysis plants have capacity in this range. The investment costs for equipment in this capacity range are manageable for small and middle-size enterprises; the operation of plants ensures countable business. However, these plants are still too small to cover the raw material demand of a big chemical company to a significant extent (10 % or more).
- 3. More than 30 000 tpa: this is a capacity range that is relevant for the chemical industry. Since 2019, the number of pyrolysis plants in this capacity range has been clearly increasing.
- Operating company/consortium:
- 1. Inventor- or founder-led companies, start-ups, or spin-offs: most operators of pyrolysis companies are process inventors or developers who try to commercialize the process. The capacity of these plants does not exceed 20 000 tpa; the ability to scale up the process is often unclear.
- 2. Waste management companies (Indaver, PHJK, Agrob Eko): waste management companies recognize the potential of chemical recycling and are already working on technical solutions, though the general conditions (oil price, waste price) are not optimal.
- 3. Equipment manufacturers, engineering companies (Splainex, Leitner Technologies, Jinan Niutex, Xinxiang Huayin):

these companies sell modular pyrolysis plants with capacities up to 35 000 tpa. A greater capacity can be achieved by numbering up.

- 4. Petrochemical companies (MOL, Indian Oil Corporation, Neste, OMV, Chevron Phylips, BP, Shell): petrochemical companies operate demonstration plants for proof-of-concept and for checking economic feasibility. However, BP (for Gelsenkirchen) and Chevron Phillips (for Baytown) have announced plans for pyrolysis plants with capacities above 100 000 tpa for PE production.
- 5. Big chemical companies (BASF, DOW, INEOS, Lyondell-Basell, Borealis, Lucite, SABIC): though these companies have know-how and the capability to develop their own pyrolysis plants, cooperation with smaller technology providers seems to be more attractive. Thus, INEOS has started a joint project with Indaver, Green Mantra, und Pyrowave; BASF has announced a cooperation with Pyrum und Quantafuel.

The economic aspects of pyrolysis plants have been discussed by the Boston Consulting Group in the report "A circular solution to plastic waste" [33]. A simplified consideration of operating costs (raw materials, plant operation) for a pyrolysis plant (30 000 tpa capacity) leads to an equivalent (break-even) crude oil price as a benchmark for commercial operation in different regions (see Tab. 2).

Tab. 2 reveals the economic "weak point" of pyrolysis plants, which focus on pyrolysis oil as the main product. Here, the price of pyrolysis oil will be directly compared with the price of crude oil of naphtha, which is highly volatile. For example, the price of oil in 2020 was on the level of 50 US\$/bbl, and the operation of pyrolysis oil was not commercially attractive. However, in 2022 the price of oil is well above 100 US\$/bbl.

A detailed description of investment in the construction and operation of a European pyrolysis plant with a capacity of 120 000 tpa can be found in [34]. This analysis, too, shows that the commercial success of such a project depends on the price of crude oil.

2.2 Solvolysis

The decomposition of polymers into oligomers or monomers in solvents under the action of these solvents or of the chemical reagents dissolved in them (bases, acids, alcohols, etc.) is called solvolysis. In contrast to pyrolysis, which can be used to depolymerize all polymers and polymer mixtures, solvolysis is a much more selective recycling process and must be adapted to

 Table 2. Operating costs for a pyrolysis plant (30 000 tpa) and equivalent oil price.

Location	Plastic waste [US\$/ton]	Cost for pyrolysis [US\$/ton]	Disposal, transport [US\$/ton]	Total cost [US\$/ton]	Equivalent oil price [US\$/bbl]
Singapore	169	196	5	370	58.90
France	120	196	5	321	51.00
USA (Florida)	125	196	5	326	51.80
China (Zhejiang)	200	196	5	401	63.80

the polymer. Since the polymer is broken down into components "step by step" during solvolysis, this technology is employed almost exclusively for polymers that are formed by polycondensation and contain heteroatoms (e.g., N or O) in the main chain. Depending on the reactant used, a distinction is made between glycolysis (e.g., with ethylene glycol, see Eq. (1) [35]), aminolysis (e.g., with ethanolamine, see Eq. (2) [36]), acidolysis (with acids), alcoholysis (e.g., with methanol , see Eq. (3) [37]), and hydrolysis (water, see Eq. (4) [38]).

Polyethylene terephthalate(PET) + ethylene glycol
$$\rightarrow$$

bis(2-hydroxyethyl terephthalate)(BHET) (1)

Polyethylene terephthalate(PET) + ethanolamine \longrightarrow bis(2 - hydroxyethyl) terephthalamide(BHETA) (2) Polyethylene terephthalate(PET) + methanol \longrightarrow dimethyl terephthalate(DMT) (3)

$$- [R_1 - O - CO - NH - R_2 - NH - CO - O_n](PU)$$

+ H₂O $\longrightarrow n NH_2 - R_2 - NH_2 + 2n CO_2 + 2n R_1OH$
(4)

The solvolysis is carried out at temperatures between 150° and 300° C; the process can be accelerated by catalysts (zeolites, bases, etc.). The exact composition of the solvolysis products depends essentially on the raw material. After separation from the solvent, these products can be used to produce the original polymer or as raw materials in the production of other chemical products. An overview of the current solvolysis plants and projects is given in Tab. 3.

Table 3. Overview of the developers of solvolysis plants, projects, and operators.

Company	Country	Process	Polymer	Products	Partner	Status/summary
ICI PLC		Glycolysis	PU	Polyols		ICI was sold to Akzo Nobel in 2007 and restructured.
H&S Anlagen- technik GmbH	DE	Acidolysis	PU	Polyols	DOW	Technology developer for recycling PU foams. Phthalic acid is used in the acid- olysis. A recycling plant with a capacity of 2500 tpa was built in Dendo, Poland. Together with DOW Polyurethanes, an additional plant is planned for the site of Orrion Chemicals Ograform in France [39]
Getzner Werkstoffe	А	Glycolysis	PU	Polyols		Getzner Werkstoffe operates its own recycling plant for the use of production waste [40].
BASF	DE	Glycolysis	PU	Polyols		BASF put a plant for depolymerizing PU into operation in Ludwigshafen in 2020. The polyols are used as secondary raw material in the production of PU.
Emery Oleo- chemicals	USA	Glycolysis	PU	Polyols		This technology combines ozonolysis with the glycolysis technology developed by INFIGREEN [41].
Polymer Research Technologies	CAN	Glycolysis	PU, PET	Polyols		The Canadian start-up company developed its own technology for transforming PU and PET for polyols for the synthesis of PU.
International Automotive Com- ponents Group North America LLC	USA	Glycolysis	PU	Polyols		Large supplier for the automotive industry with own solvolysis-based technology.
Toshiba Corporation	JPN	Aminolyse	PU	Polyols/ oligomer amines/ 1-(2-hy- droxyethyl)-oxazo lidon		Toshiba developed a continuous process. The products are used in the synthesis of PU and to produce epoxy formulations [42].
Kobe Steel, Ltd.	JPN	Hydrolysis	PU, PP	Monomers		The process using supercritical water is suitable for several polymers.



Company	Country	Process	Polymer	Products	Partner	Status/summary
Isuzu Motors Limited	JPN	Aminolysis	PU	Polyols	:	
LOOP Industries	USA	Alcoholysis	PET	Dimethyl tereph- thalate (DMT), ethylene glycol (MEG)	Indorama	The polymer is completely depolymer- ized by a solution of Na-methanolate in methanol [43].
Garbo Srl.	Ι	Glycolysis	PET	Bis(2-hydroxyethyl terephthalate (BHET)		The reprocessing specialist developed the ChemPET process for recycling clean and contaminated PET. The intermediate BHET can be used after separation to produce new PET.
gr3n	СН	Glycolysis	РЕТ, РА	Phthalic acid, MEG	Neogroup	The start-up company developed a microwave reactor for ultrafast glycolysis using ethylenglcol catalyzed by NaOH.
ioniqa	NL	Glycolysis	PET	Phthalic acid, MEG	Unilever, Indorama	The spin-off from TU Eindhoven started operation of a 10 000 tpa PET recycling plant in 2019, producing material with food-contact quality [44].
Rittec Umwelt- technik GmbH	DE	Glycolysis	PET	Phthalic acid, MEG	Reclay Systems GmbH	The company developed revolPET [®] und revolTEX [®] technologies and built a pilot plant based on old patents.
Aquafil	Ι	Hydrolysis	PA, PET	Monomers, econyl [®] -fibres		Depolymerization of fishing nets in supercritical water to produce textiles (econyl [®]).
DSM	NL	Aminolysis	PA	Monomers		DSM is a leading producer of PA 6 and PA 66 and developed a depolymerization technology for these polymers.
Meltem Kimya	TR	Glycolysis	PET	MELPET [®] rPET resins		Meltem Kimya has operated a depoly- merization plant with a capacity of 50 000 tpa for its own needs since 2014; an additional plant is scheduled for 2022 with 100 000 tpa [45].
Guangzhou Jutian Chemical Industry Technology Co., Ltd.	CN	Alcoholysis	PU	Polyols		Continuous process using microwave- supported depolymerization of PU to polyols.
Shanghai Hecheng Polymer Technol- ogy Co., Ltd	CN	Alcoholysis	PU	Polyols		The technology is a source of polyols for use in producing PU adhesives and elastomers.
IBM	USA	Glycolysis	PET	Phthalic acid		IBM markets DBU (1,8-diazabicy- clo[5.4.0]undec-7-ene) as an organic catalyst for glycolysis running at 200 °C. The process is suitable for contaminated PET.
Tyton BioSciences	USA	Alcoholysis	PET			The process operates at temperatures below 200 °C in the presence of NaOH or KOH to recover PET from textiles [46].

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Table 3. Continued.

Company	Country	Process	Polymer	Products	Partner	Status/summary
Troy Polymers	USA	Glycolysis	PU	Polyols		Troy Polymers, Inc. (TPI) is a technol- ogy developer in the field of PU and oth- er polymers. The glycolysis is supported by hydroxides and alkoholates of alkaline metals [47].
Shaw Industries' Evergreen Nylon Recycling	USA	Aminolysis	РА			Shaw Industries' Evergreen Nylon Recycling (ENR) is the world's largest Nylon-6 recycling plant and recovers 12 000 tpa. The technology originates from the Honeywell/DSM joint venture (Evergreen); however, the plant was shut down several times between 2001 and 2016.
Mura Technology	UK	Hydrolysis	PE, PP	Liquid HCs	DOW, ReNew ELP, Licella	Mura Technology markets HydroPRS TM (Hydrothermal Plastic Recycling Solu- tion) recycling technology, which was initially developed by Licella. The first plant with a capacity of 20 000 tpa will be built together with ReNew ELP; an additional plant is planned together with DOW. Mura's objective is to build recycling plants with a total capacity of 1 Mn tpa by 2025 [48].

In contrast to most pyrolysis plants, industrial solvolysis plants can be operated with a specific type of plastic waste. These are essentially polyamides (PAs), polyurethanes (PUs), and polyethylene terephthalate (PETs). PE and PP are recycled only in hydrolysis plants that use supercritical water. Glycolysis and alcoholysis or methanolysis is preferably applied in recycling PU foams and PET, while the aminolysis-based process developed by DSM is employed to recycle PA. It also becomes clear that solvolysis-based recycling processes are being developed and implemented worldwide. Most of the companies listed in Tab. 3 already operate their own plants, or they supply technology to their partners. These plants can be classified according to size, raw material, and product quality and can be divided into different groups.

Raw material:

- 1. PU: recycling either postconsumer PU foams (mattresses, upholstery) or production waste
- 2. PA: mainly PA 6 from technical applications, textiles, fishing nets, carpets
- 3. PET: packaging waste, food-packaging waste, PET fibers Product quality:
- 1. Pure monomers: phthalic acid, phthalates, and ethylene glycol from PET recycling
- 2. Oligomers and polymers: polyols from PU recycling, PET resins
- 3. Mixture of liquid-phase products: products of hydrolysis by supercritical water

Plant capacity:

1. Up to 1000 tpa: most of the solvolysis plants; the capacity already allows commercial operation

- 2. 1000–10 000 tpa: solvolysis plants in this capacity class demonstrate an important development stage for further commercialization of the technology
- 3. More than 10 000 tpa: the newest plants just constructed or currently in planning stage. The number of solvolysis plants with such capacity has been increasing since 2019.

In the past, big chemical companies (BASF, DOW, DSM) have developed their own solvolysis processes, which have not been commercialized. This tendency seems to be changing with the newest projects of DOW and BASF.

3 Impact Assessment

Mechanical recycling of plastic waste is the most economical waste-treatment strategy under almost all conditions. This can be demonstrated by life-cycle analysis. For the most important parameter, greenhouse gas emissions, the expenses for collection, transport, and processing are clearly offset by the credits from the avoided use of fossil raw materials. The net savings (expenses - credits) are in the range of 1.0-1.3 kg CO2eq kg [49]. The thermal treatment of plastic waste can be equal to or even more advantageous than recycling if the incineration plant enables the energy contained to be used efficiently to produce heat and electricity, thus substituting for fresh fossil fuels for electricity and heat generation [50]. This advantage disappears when the energy mix shifts towards renewable energies. But even then, incineration may be a better alternative for heavily mixed waste or sorting residues from plastic-waste processing.

Pyrolysis processes are a way of closing carbon cycles. If the products are used as fuel only, it is not possible to approve them as recycling processes [51]. The requirements for the purity of the input material are lower than with mechanical processes. Highly heterogeneous mixtures of substances and composite materials can also be processed in solvolysis processes [8].

Chemical recycling requires more effort than mechanical recycling and is therefore not in competition with it. Whether chemical recycling has advantages over thermal treatment cannot be decided yet with certainty, due to a lack of large-scale technical experience. While it is sufficient for mechanical recycling to sum up the costs for transport and processing in energy (or exergy) per kg of plastic waste (see, e.g., [52]), in chemical recycling, all the substances used (hydrogen, solvents, additives such as CaO) must be considered [14].

A comparison of four studies on chemical recycling sees lower greenhouse gas emissions than with incineration (even taking energy credits into account), but only if the required hydrogen can be produced without releasing CO_2 [53]. This is certainly true as long as chemical recycling plant capacities are still small. At some point, however, chemical recycling will compete with other green hydrogen customers, such as the steel industry or possibly the transport sector. Such a competitive situation has been described in regard to electricity generation for electromobility (merit order approach, [54]). Environmental organizations criticize eco-balances that see clear advantages in chemical recycling [55].

Use of CO_2 as a raw material:

Besides the energetic aspect of the effective thermal use of plastic waste, which will increasingly experience ecological competition from hydrogen as an energy source, a large part of today's carbon cycle is found in organic compounds; here, pure hydrogen products cannot be alternatives. The plastics themselves, along with the synthetic fuels, create the demand for such hydrocarbon compounds. The requirements for plastics are so specific that even the smallest impurities influence the physical and thus also the mechanical, thermal, and a number of other properties.

Mechanically recycled plastics thus require a very high level of effort to control the input material for further use. In most cases, the effort and thus the costs of combining several mechanical processing steps must be weighed against the sales proceeds of the recyclate as a benefit. In many cases, this calculation is not (yet) positive, since compliance with increasingly stringent limits on trace substances can also almost preclude the use of recyclates (e.g., of bromine compounds in flame retardants).

The alternative could therefore be processes that both thermally use the plastic streams and provide the resulting (pure) CO_2 as a raw material for syntheses with green hydrogen. Such processes molecularize the plastics and are highly tolerant of interfering substances or inseparable mixtures of substances. Attention must be paid to the costs of CO_2 capture, as fossil carbon sources will cease to exist in the future and thus certain carbon flows will be missing.

While the incineration of plastics is also a chemical process, it is not generally associated with recycling. However, plants using combustion in an oxygen atmosphere or at least with enriched oxygen (oxyfuel) offer the possibility to more fully integrate processes for CO_2 capture after thermal treatment and to provide for better efficiency and optimized process characteristics [56].

Recirculating carbon is therefore possible. Persistent organic pollutants ((POPs), e.g., polychlorinated biphenyls (PCBs), dioxins and furans, brominated compounds) are comprehensively destroyed, but so is the polymer structure.

The future challenge will be to find and shift the dividing point dynamically between the unacceptably high use of energy for sorting and material plastic use and the demand for carbon. The processes listed under chemical recycling in Fig. 2 have equal importance.

4 Conclusion

During pyrolysis or gasification, liquid and gaseous products are obtained that can be used to synthesize plastics or at least as fuels. However, the polymer structure is not preserved here, either. The processes are therefore not considered material recycling within the meaning of \$16 of the German Packaging Act (VerpackG) [51,57]. The rate of the mechanical recycling of composite packaging is currently 55 % and will be increased to 70 % by 2022. The same applies to solvolysis, which is suitable for PAs and Pes.

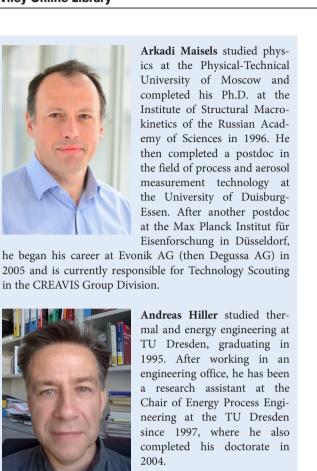
The process-engineering effort is greater in chemical recycling than in mechanical recycling. The extent to which this effort can be made wherever plastic waste is generated that cannot be mechanically recycled is questionable. In countries with a highly developed chemical industry, however, this should be achievable. Plants with a very large capacity could then also be built and operated there by importing additional quantities of plastic waste. The locations of the processes listed in Tabs. 1 and 2 are predominantly in such countries.

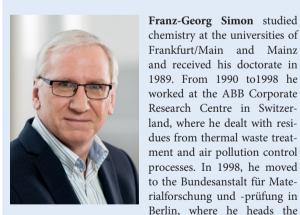
Detailed energy and mass balances and proof of industrial suitability are required to assess whether chemical-recycling processes have ecological advantages over incineration or mechanical recycling. Then, the future position of chemical recycling in the waste hierarchy could be redefined [51]. Further intensive research and development for efficient processes and concepts in chemical recycling are therefore necessary [58].

Conflicts of Interest

The authors declare no conflict of interest.







Division of Contaminant Transfer and Environmental Technologies.

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Abbreviations

ABS	acrylonitrile butadiene styrene
BHET	bis(2-hydroxyethylene terephthalate)
BHETA	bis(2-hydroxyethyl) terephthalamide (BHETA)
DMT	dimethyl terephthalate
HC	hydrocarbon
HDPE	high density polyethylene
LDPE	low density polyethylene
MEG	ethylene glycol
MMA	methyl methacrylate
PA	polyamide
PE	polyethylene
PET	polyethylene terephthalate
PMMA	poly(methylmethacrylate)
PP	polypropylene
PS	polystyrene
PU	polyurethane
PVC	polyvinyl chloride
WEEE	waste electrical and electronic equipment recycling

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