2014 - 2020 Interreg V-A
Italy - Croatia CBC Programme
Call for proposal 2019 Strategic

MARLESS (MARine Litter cross-border awarenESS and innovation actions)

Priority Axis: Environment and cultural heritage; Specific objective: 3.3 - Improve the environmental quality conditions of the sea and coastal area by use of sustainable and innovative technologies and approaches

6.2.1 – Report: status of the art on thermochemical treatments of plastic marine litter. Comprehensive analysis of the state of the art of the different international projects dealing with the treatment of plastics in shallow and deep waters

WP6 – Action 6.2

Version: FINAL
Distribution: PUBLIC
Date: 31/08/2021

PROJECT MARLESS

Work Package:	**WP6 - Pilot Actions to prevent, recover, process marine litter, including Stakeholders Toolbox**
Activity:	**6.2 - Pyrolysis micro plant in Ravenna: test what we do**
WP Leader:	**Alma Mater Studiorum Università di Bologna**
Deliverable:	**6.2.1 – Report: status of the art on thermochemical treatments of plastic marine litter. Comprehensive analysis of the state of the art of the different international projects dealing with the treatment of plastics in shallow and deep waters**

Version:	**Final 1.0**	**Date:**	**31/08/2021**
Type:	**Report**		
Availability:	Public		
Responsible Partner:	Alma Mater Studiorum Università di Bologna		
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INDEX

ABSTRACT AND SCOPE

Within the last decades, plastics have revolutionised our daily life. Heavy materials have been substituted with plastics, making their application easier, such as in the case of medical devices. Plastics, however, shows a dark side: being a robust material, its lifetime is almost infinite, and this means that its end-of-life disposal is important to avoid environmental problems. This report starts with a brief introduction about plastics and marine litter. Then, the ways available to dispose and/or recycle plastic are described, with a particular focus on thermochemical processes, and more specifically on pyrolysis, the thermal decomposition of materials at high temperature in the absence of oxygen. Finally, an overview is given of the most important European projects dealing with plastic disposal and recycling challenges. Alongside these European projects, which mainly involve public bodies such as Universities and Public institutions, private companies, such as Yamaha and LyondellBasel, are focusing on the same theme. Everyone's aim is the same: to find a solution about plastics end-of-life that could be environmentally friendly, sustainable and profitable. The scope of this work is to give an overview on the technologies available and to explain how the work foreseen in MARLESS could help in this scenario.

1. INTRODUCTION

Plastics are a diverse group of synthetic polymers that have their origins in the late 19th century, when Leo Baekeland first synthetized them. However, rapid growth in global plastic production started in the 1950s. These polymers can be divided in 2 main categories: thermoplastics and thermosets. The primary difference between the two is that the former can be reheated, remoulded, and cooled as necessary without causing any chemical change, the latter is a material that strengthens when heated, but cannot be remoulded or heated after the initial forming. Thermoplastics contribute to the total plastic consumption by roughly 80% and are used for typical plastics applications such as packaging but also in non-plastics applications such as textile fibres and coatings. Production of plastics is growing at a rate of about 9% per year worldwide, with nearly 370 million tons in 2019 (see Figure 1). Plastic's end-of-life management is nowadays a hot environmental topic (Our World in Data 2021).

Plastics diffusion in our daily life has been possible because they are a good solution to our fast-changing needs, due to their durability, low production cost, light weight and the possibility to fulfil numerous requirements, such as formability, heat resistance and insulation (Davis 2019; Taylor and Kennedy 1994). Beside our daily life, several sectors have benefited from the development of different plastic kinds. For example, they revolutionized medicine with life-saving devices, made space travel possible, lightened cars and jets, thus saving fuel and pollution, and saved lives with helmets, incubators, and equipment for clean drinking water. However, plastic's durability means that it persists in the environment for many years, and its low density means that plastics are readily dispersed by water and wind. Moreover, most of the plastic used today is for single use, such as food wrappers and plastic bags, with a lifespan of mere minutes to hours but they may persist in the environment for hundreds of years. Single-use plastics account for 40 percent of the production every year, being so a tricky environmental problem (Chen et al. 2021). Since 1972, it has become clear that a wrong disposal of plastics negatively impacts the environment due to long degradation time, and so research on plastic management and recycling started with the creation of the first plastic waste recycling mill in Pennsylvania (Pohjakallio 2020). The exponential growth of plastic production in the last 50 years implies that on the one hand, more resources are being used to meet the increased demand of plastic, and on the other hand, more plastic waste is being generated.

Changing the waste into valuable useful resource might be a way to make use of the waste with the goal to meet the increasing power demand.

To summarize, plastics can be divided in seven main groups, as it can be seen in Table 1.

Polymer	**Acronym**	**ID code**	**Applications**
Polyethylene terephtalate	PET	1	Plastic bottle production
High-density polyethylene	HDPE	2	House wrap, bottle crates, toys and playground equipment
Polyvinyl chloride	PVC	3	Pipes, electric cables, construction, signs, clothing, health care
Low-density polyethylene	LDPE	4	Containers, plastic wraps, playground slides
Polypropylene	PP	5	Clothing, medical, niche
Polystyrene	PS	6	Packaging, household appliances, building and construction
Other		7	Other

Table 1. Plastic materials classification and their application

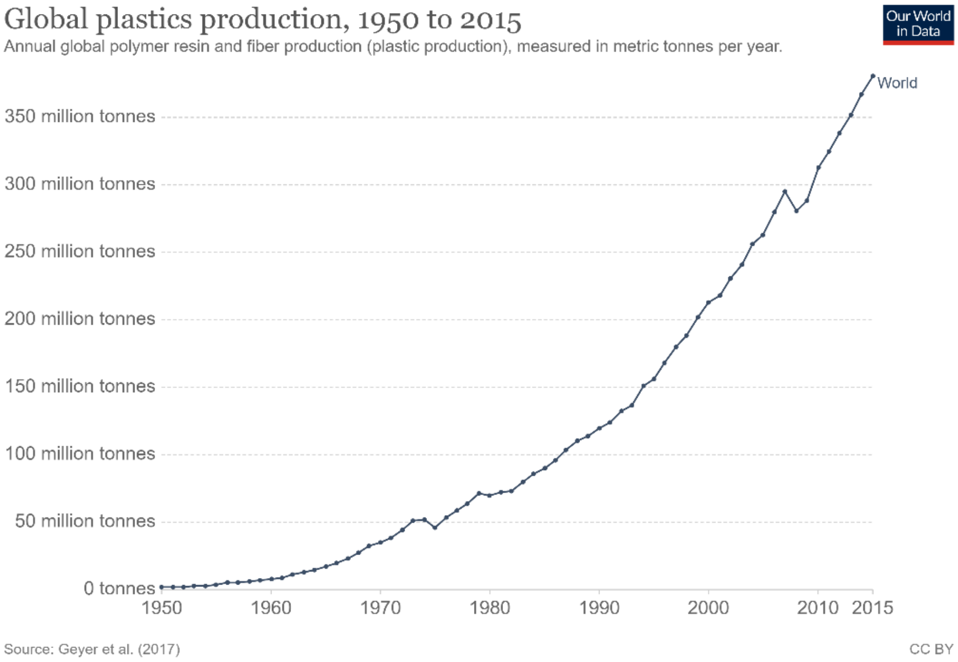

Figure 1. Global plastic production from 1950 to 2015. (Our World in Data 2021)

Every year, about 8 million tons of plastic waste escapes into the oceans from coastal nations. This leads to severe environmental and ecological problems, especially because of the presence of additives, which are used to make the material stronger, more flexible, and durable (Hahladakis et al. 2018). On the other side, this means that their life and persistence is extended, with some estimates ranging to at least 400 years to break down. This means that all the plastic ever produced still exists somewhere in the biosphere, although much of it is now invisible to humans, reduced to tiny particles in the ocean and land ecosystems (microplastics). Figure 2 shows how many years are needed for different marine items to decompose. All these problems could be solved in two different ways, either by replacing plastics with alternative materials or through recycling. However, it has been assessed that plastic replacement with other alternative materials requires 22.4 million extra tons of crude oil when we take into consideration a life cycle perspective. Plastic recycling seems to be the most feasible solution, even if, also in this case, environmental and economic variables should be considered (Milios et al. 2018; Schyns and Shaver 2021).

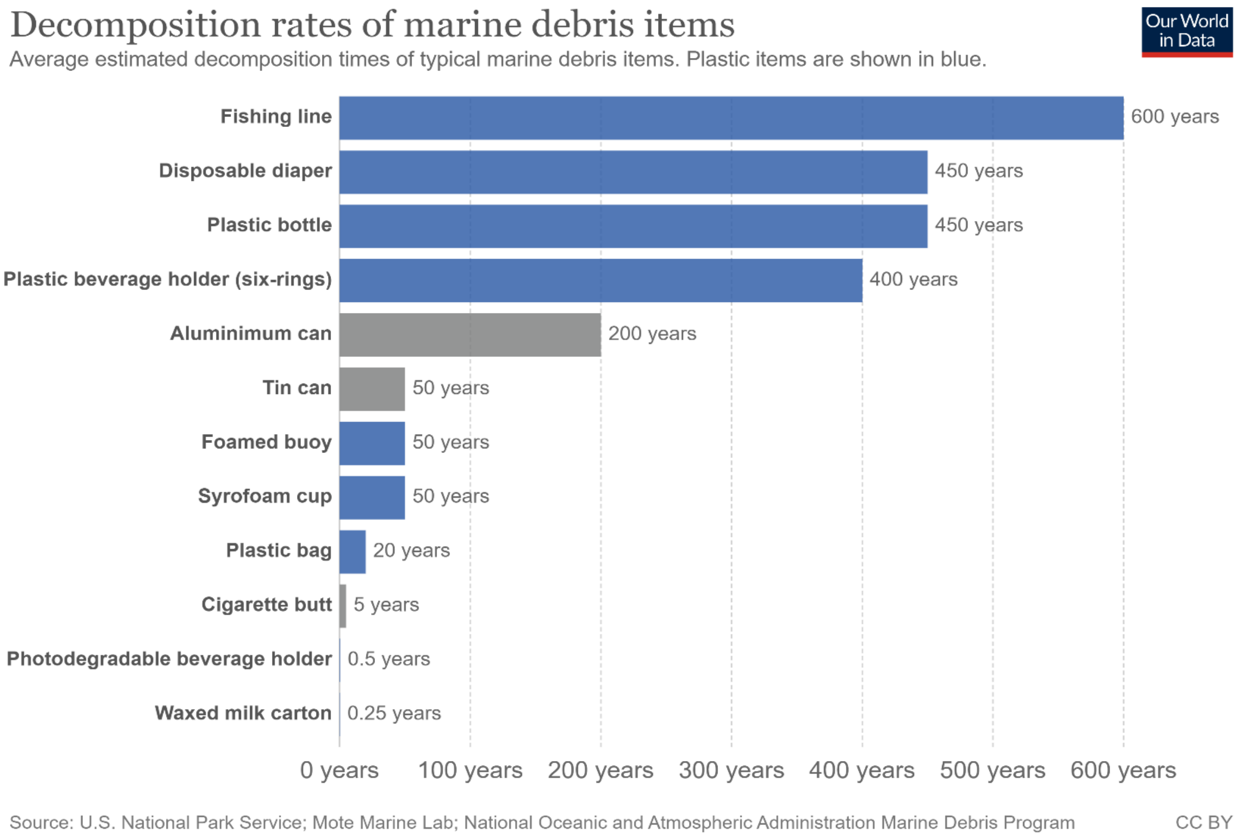

Figure 2. Decomposition rates of marine debris items. (Our World in Data 2021)

2. MARINE LITTER

According to the United Nations Environment Programme, marine litter is defined as “any persistent, manufactured or processed solid material discarded, disposed of or abandoned in the marine and coastal environment” (UNEP - UN Environment Programme 2017). Today, marine litter is recognized as a major concern for the future of the seas and consists of a wide range of materials, including plastic, metal, wood, rubber, glass and paper. Although the relative proportions of these materials vary regionally, with different chemical composition and physical characteristics, there is clear evidence that plastic litter is by far the most abundant type. In some location, plastics make up 90 % of marine litter in shorelines. Plastics also deteriorate and fragment in the environment because of the exposure to sunlight (photo-degradation) in addition to physical and chemical deterioration. The breakdown of larger items results in numerous tiny plastic fragments, which, when smaller than 5mm are called secondary micro plastics. Other micro plastics that can be found in the marine environment are categorised as primary micro plastics because they are produced either for direct use, such as for industrial abrasives or cosmetics or for indirect use, such as pre-production pellets or nurdles. Commonly found plastics in the oceans and seas are cigarette butts, bottle cups, straws, cups and plates, single use bags, food wrappers and beverage bottles. Marine litter management is challenging because the material is not only very heterogenous but also strongly weathered by various environmental influences, such as mechanical degradation based on wave movement and sandy shores as well as degradation due to UV radiation, oxidation and the general process of biofouling.

Based on these characteristics and the size of the fragments, marine litter may accumulate in different abiotic and biotic matrices: sea surface, water column, shoreline, seafloor, sea ice and biota. The Mediterranean Sea is one of the most affected basins by marine litter worldwide, and this has impacts on vital economic sectors such as fisheries, aquaculture, navigation, energy, and tourism, other than endangering human health and safety (Laist 1997). Another serious problem is the entanglement of fish and dogfish in rubber bands, which was reported by Anon (1971) and Gochfeld (1973), who highlighted the entanglement threat posed by marine litter to coastal birds. Litter monitoring, which means repeated surveys of beaches, seabed, water column, surface waters and biota to determine litter types and quantities in a representative manner such that information can be compared with baseline data to follow trends, is an indispensable action to understand how to act to fight this problem. Movements of plastics in the water, both in the vertical and in the horizontal dimension, depend on the characteristics of plastic waste and the local environmental factors. The vertical movement of plastics in water, or sedimentation rate, is mostly affected by three characteristics: the density, the surface area of the polymer, and the particle size (Erni-Cassola et al. 2019). Horizontal movements are determined by wind and currents, which may accumulate large quantities of plastics in very specific places (Thushari and Senevirathna 2020).

About the kind of plastics in the ocean, the most frequent is polyethylene, which is dominant in most environmental compartments, followed by polypropylene and polystyrene. Studies have demonstrated that in open oceans, microplastic polymer types segregated in the water column according to their density. Lower density polymers, such as polypropylene and polyethylene, dominated sea surface samples (25% and 42%, respectively) but decrease in abundance through the water column (3% and 2% in the deep-sea, respectively), whereas only denser polymers (such as polyesters and acrylics) were enriched with depth (5% in surface seawater vs.77% in deep-sea locations).

In the environment, plastic is known to deteriorate and fragment therefore occurring in a wide range of sizes, for which standardized categories have very recently been proposed: macro (≥1 cm), meso (1–10 mm), micro (1–1000 μm) and nano plastics (1–1000 nm), with smaller particles being numerically most prevalent on sea surfaces (Erni-Cassola et al. 2019; Hee et al. 2021; Khatmullina and Chubarenko 2019). In the water, however, degradation is slowed down by the low temperatures and the limited UV penetration, leading plastic debris to persist and accumulate. These studies confirm that PE, PP, PS and PP&A are among the most abundant polymer types in aquatic environments. This is not surprising as these materials accounted for 74% of global plastic production in 2015 and are commonly used in short life-cycle products.

3. PLASTIC'S END OF LIFE

After food waste and paper waste, plastic is the mayor waste coming from municipal and industrial sector and its management is challenging. In Europe, waste management was legislated for the first time in 1975 (Directive 75/442/EEC) with the aim of reducing waste production and its harmfulness. This directive has been modified through the years and waste management is currently legislated by the Waste framework directive 2008/98/EC. Plastic materials have two main destinies at the end of their life: disposal or recycling (Randall Curlee 1986). In the case of disposal, landfill and incineration are the options to be considered. In the case of recycling, mechanical and chemical processes are available. In all cases, technical and economic feasibility and overall commercial viability of the recycling method must be considered in each step of the chain. Plastic disposal in landfills may contaminate waterways and aquifers with plasticizers and other chemical additives. Moreover, plastics are not biodegradable, so that the landfill solution is undesirable because of the increasing cost and decreasing surface available (Hahladakis et al. 2018). On the other side, incineration of the plastic waste can be a major source of air pollution and the pollutants liberated from this process, especially when PVC is burnt, contribute to climate change (Verma et al. 2016). For what concerns the recycling options, the interest in chemical recycling has increased in the last years, because oil resources are diminishing and transforming plastics back to its initial state could help to partially solve the production problem. Approximately, 4% of the fossil fuels produced globally are used to manufacture plastics and another 4% is used to generate power for the plastic manufacturing industries. It has also been predicted that the production of 380 billion plastic bags and wraps yearly in the U.S. consumes 1.6 billion litres of oil (Chen et al. 2021).

The products from pyrolysis or gasification, which will be better described in the next chapter, can be used both for energy recovery and to produce monomers, so this option is in between chemical recycling and energy recovery. Energy recovery involves complete or partial oxidation of the material, producing heat, power and/or gaseous fuels, oils and chars besides by-products that must be disposed of, such as ash. The other recycling option is the mechanical one, which transforms the waste into “new” secondary raw materials without changing the base molecular structure of the material. For example, after sorting and processing, plastic is shredded, then melted down and then re-extruded into plastic pellets. However, current mechanical recycling processes are limited by cost, degradation of mechanical properties and inconsistent quality products (Al-Salem, Lettieri, and Baeyens 2009), so that this recycling option seems not to be the best choice. Moreover, it is important to remember that thermosets are characterized by cross-linked molecular bonds that prevent those resins from melting once formed, and this characteristic makes this material difficult to be recycled (Randall Curlee 1986). Because of this, mechanical recycling usually involves mainly thermoplastics.

Recycling technologies can reduce the accumulation of plastic wastes, yet they also pollute the environment, consume energy, labour, and capital cost. Finally, it has to be taken into account that nowadays more than 300 types of plastics are produced (Chen et al. 2021). This means that waste pre-treatment becomes even more challenging, especially in the case of mechanical recycling.

3.1. TERMOCHEMICAL CONVERSION PROCESSES

Thermochemical processes can be divided into advanced thermo-chemical or **pyrolysis** (thermal cracking in an inert atmosphere), **gasification** (in the sub-stoichiometric presence of air usually leading to CO and CO_2 production) and **hydrogenation** (hydrocracking) (Brems et al. 2013).

The pyrolysis process involves thermal degradation of plastics at high temperatures (usually 400°C to 800°C) in the absence of oxygen and will be better described in the following chapter.

In the gasification process, plastics is reacted with a gasifying agent at high temperatures, up to 1300°C, with the aim to produce syngas, which can be used as a building block for the production of other products or as a fuel in fuel cells (Saebea et al. 2020). Syngas characteristic can be managed adjusting some process parameters, like temperature and pressure. The studies dealing with plastic gasification are in general scarce. However, due to the knowledge acquired on biomass and coal gasification, the advance in the development of plastic gasification technologies is considerable with several gasification studies at the pilot scale unit already performed. Air gasification is the most studied and developed strategy and pursues the production of a syngas for energy purposes. In spite of the higher H_2 content and heating value of the gas produced by steam gasification, these alternative faces significant challenges, such as the energy requirements of the process and the tar content in the syngas (Lopez et al. 2018).

Hydrogenation process is based on the fact that waste commodity plastics have a high hydrogen to carbon ratio and molecular chain structures suitable for liquefaction. Therefore, direct liquefaction can be considered as a potential waste disposal option by generating oil, which could be upgraded to transportation fuels. The nature of polymer and solvent, degradation temperature range, catalyst, reaction time, and hydrogen pressure can affect the oil yield and characteristics. Liquefaction of polymers can be treated as the thermal breakdown of the polymer accompanied by hydrogenation of the broken chain ends. If a closed system is utilized during the process, this thermochemical solution is interesting as well because in this case there are no harmful emissions to the environment.

To summarise: depending on the conversion method, plastics waste can be selectively converted either to bio-oil, bio-crude oil, synthesis gas, hydrogen, or aromatic char. Thermochemical treatment has mainly three advantages: it converts waste into safe forms (bio-oil and gas, usually), the weight and volume of waste is significantly reduced, substantial amount of heat can be recovered and recycled back to the process. Incineration is advantageous treatment because of the heating value (LHV) of plastics and it has been shown that PE has the same LHV as diesel fuel. Holmgren (2006) describes a heating system that uses waste heat supplied from various sources, in the city of Gothenburg (Goteborg) in Sweden. She shows that heating networks can utilize waste heat that would otherwise have limited use. Incineration and on-site use are advantageous also because they eliminate the need for transportation to external (regional) waste disposal facilities/contractors and the for the consequent fuel consumption.

Thermal degradation processes allow obtaining several constituting molecules, combustible gases and/or energy, with the reduction of landfilling as an added advantage (Mastral, Berrueco, and Ceamanos 2007). The main thermochemical treatments of waste plastic are summarised in Table 2.

Process	Products
Steam gasification	Syngas
Air gasification	Energy
Pyrolysis	Fuels and chemicals
Hydrogenation	Fuels and chemicals

Table 2. Thermochemical conversion processes of plastic waste and their outputs (Lopez et al. 2018)

3.2. PYROLYSIS OF PLASTICS

Pyrolysis is the process of thermally degrading long chain polymer molecules into smaller, less complex molecules at high temperature, generally 400-800°C, and in absence of oxygen. The process output consists in oil, gas and char and can take place both in the presence or absence of a catalyst. One of the advantages of this process is that all the products have a commercial value: gas can be recycled to feed the process heat, oil can be refined, if needed, to produce advanced fuels or chemicals, and char can have many applications, especially in the concrete industry. Since plastic has a high calorific value, the calorific value of the gas is high as well, with a value of 22–30 MJ/m^3 depending on the waste material being processed (Fivga and Dimitriou 2018). Pyrolysis provides several other environmental and operational advantages and financial benefits. Operational advantages could be described by the utilisation of residual output of char used as a fuel or as a feedstock for other petrochemical processes. An additional operational benefit is that pyrolysis requires no flue gas clean up as the flue gas produced is mostly treated prior to utilisation. Environmentally, pyrolysis provides an alternative solution to landfilling and reduces greenhouse gas and carbon dioxide emissions by substituting fossil fuels.

The process produces a high calorific value fuel that could be easily marketed and used in gas engines to produce electricity and heat. However, several obstacles and disadvantages do exist as well, mainly the handling of char produced, and the treatment of the final fuel produced if specific products are desired. Different plastic waste types influence the yield and the quality of the produced oil, so that waste pre-treatment is preferred, even if not always necessary. Fundamentally, different types of plastics have different compositions that normally is reported in terms of their proximate analysis, a technique used to measure the chemical properties of the plastic compound based on four elements which are moisture content, fixed carbon, volatile matter and ash content. Volatile matter and ash content are the major factors that influence the liquid oil yield in the process. A large quantity of volatile matter favours generally the liquid oil production, while high ash content decreases the amount of liquid oil, consequently increasing the gaseous yield and char formation.[20] Yield and composition of the resulting oil depends on the processed feedstock composition. A study made by Miandad et al. (2017) showed that PS plastic waste showed maximum production of liquid oil (80.8%) along with least production of gases (13%) and char (6.2%) in comparison to other plastic types. Liquid oils from all plastic types contain mostly aromatic compounds with some alkanes and alkenes. Liquid oil from PS pyrolysis contained styrene (48.3%), ethylbenzene (21.2%) and toluene (25.6%). Oil characteristics, which are summarized in Table 3, have been shown to be very similar to conventional diesel, therefore the pyrolysis output has good potentials as an alternative energy source for electricity generation.

Property	Value
Dynamic viscosity	1.77 - 1.90 mPa s
Kinematic viscosity	1.92 - 2.09 cSt
Density	0.91 - 0.92 g/cm^3
Pour point	11 - 60°C
Freezing point	15 - 65°C
Flash point	28.1- 30.2°C
High heating value	41 MJ/kg

Table 3. Pyrolysis oil characteristics (Miandad et al. 2017).

Upgrading of liquid oil using post-treatment methods such as distillation, refining and blending with conventional diesel is required to make it suitable as a transport fuel due the to presence of high aromatic compounds. The recovery of aromatic compounds, especially styrene, from pyrolysis oil can be a potential source of precursor chemical in industries for polymerization of styrene monomers. However, there is not a sufficient understanding of the underlying reaction pathways, which has prevented a quantitative prediction of the full product distribution. Both batch and continuous plants can be used. In a waste management company, where the aim is to recycle most of the waste and to have a good economic benefit, continuous plants are preferred.

4. WHAT HAS BEEN DONE TILL NOW AND RESULTS

Awareness raising resulting in behavioural change, especially among young people, is an integral part of any meaningful strategy against marine litter. It includes the understanding that prevention is the only long-term solution to pollution and that the contribution of everyone, according to competences and capabilities, is necessary. The EU not only finances dedicated projects focused on awareness raising but also requires dissemination and communication activities in almost all EU-funded projects against litter. They focus on prevention or reduction of marine litter through a range of actions at national or regional level such as improved waste and wastewater management, port reception facilities, targeted fishing for litter, education, awareness raising and outreach activities. The EU participates actively in the development and implementation of these action plans and promotes their coordination. An example of EU-founded project is LIFE - MERMAIDS[1] ("Mitigation of microplastics impact caused by textile washing processes"), which lasted 2 years, from 2014 to 2016, and which had the objective to contribute to the mitigation of the environmental impacts on European marine ecosystems of microplastics found in laundry wastewater. The project demonstrated and implemented innovative technologies, such as additives for detergents and textile finishing treatments, to prevent microplastic removal from synthetic clothing during the laundry processes. In order to contribute to real design concepts and industrial application, four pilot study sites with different environmental characteristics were identified: Baltic sea, North Sea, Atlantic Ocean and Mediterranean Sea. The sites represent specific challenges in relation to environmental, social, and economic conditions. Reductions in microplastic release in the range of 10%-70% were shown to be possible (though high variability was noted), when specific detergents and textile additives were used in low temperature / low cycle laundries. The projects evaluation of environmental benefits confirmed a reduction in microplastic release of about 23% for a domestic laundry and 7% for an industrial laundry.

Another interesting project, which ended in December 2020, is the EU project marGnet[2] ("Mapping and recycling of marine litter and ghost nets on marine floor"), which aimed at setting up and testing multi-level solutions to monitor, map, prevent, remove and recycle marine litter from sea-based sources present on the sea-floor. It is often assumed that marine litter floats in oceans, however part of the litter ends up on the seafloor, especially that generated by the fisheries and aquaculture activities. In Europe, these materials account up to 27% of all marine litter. The project has been carried out with a holistic approach, combining actions to tackle the phenomenon of marine litter at

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[1] https://www.life-mermaids.eu
[2] https://www.margnet.eu

all phases, from reduction and prevention, through the monitoring and quantification and the removal and recycling through thermochemical treatments for fuel production. A portable prototype capable of transforming plastic waste present in the sea in fuel for boats, at a low cost, through a pyrolysis process at low temperatures has been developed by the Institute of Marine Sciences of the National Research Council of Venice (CNR-ISMAR). The project worked on two pilot sites located in the Northern Adriatic and the Cres-Losini Archipelago in Croatia, which have two types of sea floor, so a range of field activities have been performed in both sandy and rocky bottoms, as well as in coastal and lagoon areas. This approach improves the robustness and the replicability of the project. Results indicate that, compared to the conventional marine diesel, using marGnet fuel would save approximatively 0,75 t of CO_2 per tonne of fuel. The great advantage of this process is given by the possibility of using marine waste recovered from the seabed without the need for special pre-treatments, which in fact undermine the economic sustainability of the mechanical recycling solutions attempted so far at an international level (Consiglio Nazionale delle Ricerche 2021).

4.1. INTERNATIONAL ONGOING PROJECTS DEALING WITH THE TREATMENT OF PLASTICS

Several EU and private research projects are active with the goal of reducing and recycling plastic waste. At present, more than 25.8 million tonnes of plastic waste are produced per year in EU Member States, with only 29.7% being recycled. In the next chapters, some of the main running projects will be presented. Several factors and variables need to be considered to evaluate the best option for plastic waste management and recycling. First, reduction of waste production is the best option, and this could be done by reusing most of the plastic items present in our daily life. Plastic bags itself can be used several times before disposal, even if they are used mainly as single-life items and disposed immediately after use. When the material cannot be longer reused, its disposal has to be done in the correct way to allow recycling and to avoid expensive and time-wasting pre-treatment processes. Dispersion of the materials in the environment should be strictly prohibited, but there are many developing countries that do not have a specific legislation about this issue. The consequence is that a lot of plastic reaches rivers and sea, causing severe environmental problems. A proper knowledge of plastic distribution in waters, their transport and their composition is necessary to face the problem and find solutions. At the end, the collected plastics can be recycled either mechanically or chemically. All these aspects are considered by the research groups involved in various projects, which are described in the next sections and summarised in Table 4.

Title	EU financing line	Years	Main Activity	Web site
COMMON	European Neighbourhood Instrument - Cross-Border Cooperation in the Mediterranean	2019-2022	Monitoring	https://www.enicbcmed.eu/projects/common
CLAIM	H2020 (GA no. 774586)	2017-2021	Data modelling, technological development	https://www.claim-h2020project.eu
TOPIOS	European Research Council Starting Grant	2017-2022	Data modelling	https://topios.org
PLASTICIRCLE	H2020 (GA no. 730292)	2016-2020	Technological development	https://plasticircle.eu/home/
MAELSTROM	H2020 (GA no. 101000832)	2021-2024	Technological development	https://www.maelstrom-h2020.eu
CLEANSEA	LIFE (LIFE 15 GIE/IT/000999)	2016-2020	Increase awareness	http://www.parcoasinara.org/it/contenuti/articoli/dettagli/521/
SOUPLESS	LIFE (LIFE17 ENV/NL/000339)	2018-2022	Technological development	https://webgate.ec.europa.eu/life/publicWebsite/index.cfm?fuseaction=search.dspPage&n_proj_id=6745
WES	European Neighbourhood Instrument - Southern Neighbourhood Region	2019-2023	Increase awareness, pollution reduction	https://www.wes-med.eu

Table 4. Most recent EU projects on marine litter

4.1.1. COMMON (Coastal management and monitoring network)

Started in September 2019, the COMMON project[3] ("COastal Management and MOnitoring Network for tackling marine litter in Mediterranean Sea") is financed through the European Neighbourhood Instrument - Cross-Border Cooperation in the Mediterranean and aims at monitoring marine litter. The project will end in September 2022 and the expected impact is an enhanced capacity of public authorities in monitoring and managing the waste chain with a focus on the land-sea relationship in 5 coastal areas: Tyre (Lebanon), Maremma and Northern Puglia (Italy) and Kuriat Island and Monastir (Tunisia). The integrated and multi-stakeholders approach promoted by the project will boost

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[3] https://www.enicbcmed.eu/projects/common

citizens' participation in marine litter removal activities, as well as the exchange of good practices and experiences among local institutions at Mediterranean level. So that, compared to MARLESS, this project is not focused on thermochemical waste treatment, but on monitoring and managing waste chain.

4.1.2. CLAIM (Cleaning Litter by developing and Applying Innovative Methods in european seas)

The EU H2020 CLAIM project[4] ("Cleaning Litter by developing and Applying Innovative Methods in european seas") will power 5 new technologies to innovate the ways in which we clean our seas and oceans. Data modelling will produce maps of concentrations of macro and micro litter, while ecosystem service approaches will identify areas where intervention has the greatest potential to secure impact on human well-being. Right after an effective pre-filtering system has sorted and collected litter, a photocatalytic nanocoating device will degrade microplastics in wastewater treatment plants. Mounted on ships a small-scale thermal treatment device (pyrolizer) will be used to turn collected litter intro energy powering ships and heating up ports. At river mouths, innovative floating boom will collect and monitor visible litter, while a CLAIM network of FerryBox systems will operate on ships in the Baltic, West and East Mediterranean with an automated seawater sampling device and passive flow-through filtering system. An ecosystems approach will guide the project through the evaluation of the potential benefit to ecosystem services and human well-being of the proposed litter cleaning methods. New business models will enhance the economic feasibility for upscaling the innovative cleaning technologies, considering the existing legal and policy frameworks in the CLAIM countries, as well as the acceptance of the new technologies by end-users and relevant stakeholders. Within the framework of CLAIM, the company New Naval Ltd. developed an innovative floating boom and marine litter collection and recovery system, the "Tactical Recovery Accumulation System Hellas" (CLEAN TRASH). The marine litter management system is an unparalleled, highly efficient solution, that actively controls, collects, and removes visible litter (mainly macroplastics) at river mouth locations before it can enter into the open sea and affect marine ecosystems. This system is designed to address several types and sizes of marine litter while utilising a highly efficient storage design that increases collection capacities using multiple layers of alternating compartments. The scalability and tailor-made configuration of the system allow its installation and operation in almost any river mouth location, making the system ideal to combat the influx of single-use plastics resulting from the coronavirus pandemic, while continuing to address the daily marine litter challenge. CLAIM is also handling the next step of combating plastic pollution – safely transferring collected litter to a

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[4] https://www.claim-h2020project.eu

treatment device, called pyrolyzer. The latter is a small-scale thermal treatment device mounted on boards and integrated into harbours, which transforms solid plastic waste into a combustible gas. The pyrolyzer can be used immediately on-site as an energy source for ships and lighting and heating in ports. The project's set of generally highly demanded innovative technologies become tools of higher necessity during and after the COVID-19 pandemic for their capacity to continuously contain and remove litter from the marine environment.

4.1.3. TOPIOS (Tracking Of Plastic In Our Seas)

The TOPIOS project[5] ("Tracking Of Plastic In Our Seas") is a 5-year (2017-2022) research project focused on developing a good understanding of the way plastic litter moves through the ocean, to help organize a proficient collection and recycling system. To achieve the goal, an innovative, powerful and comprehensive model for tracking marine plastic through the oceans will be developed.

4.1.4. PLASTICIRCLE (Improvement of the plastic packaging waste chain from a circular economy approach)

Alongside chemical recycling and energy recovery, mechanical recycling is still a solution that needs to be considered. The PlastiCircle project[6] ("Improvement of the plastic packaging waste chain from a circular economy approach") is rolling out innovations in waste collection, transport, sorting and recycling, and aims to transform plastic packaging waste into valuable products. Smart containers to increase collection rates of plastic waste, cost-effective waste transport systems connected to IoT cloud platforms, innovative optical sorting technologies to improve sorting and new value-added recycled plastic products are being developed thanks to the research team. PlastiCircle plans to reprocess the recovered materials into added-value products like automotive parts, foam boards for wind turbines and roofing structures, garbage bags, asphalt, fences and benches - based on extrusion, injection and compression moulding. The knowledge created will lead to a better valorisation of these materials within the market. The manufacturing of these products is to be based on extrusion, injection and compression moulding.

4.1.5. MAELSTROM (MArinE Litter SusTainable RemOval and Management)

The MAELSTROM project[7] ("MArinE Litter SusTainable RemOval and Management") is focused on removal of marine litter by designing two innovative technologies to remove litter in previously iden-

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[5] https://topios.org
[6] https://plasticircle.eu/home/
[7] https://www.maelstrom-h2020.eu

tified accumulation hotspots. These two technologies, an underwater cable robot and a Bubble Barrier, will be tested in the rivers of Portugal and Italy, removing litter both from the seabed and the water column and preventing it from reaching the sea. Collected litter will be recycled and put back into the market in the form of chemical precursors, polymers and materials that can be part of the industrial chain. In this way, litter will become a new resource, within a circular economy perspective.

4.1.6. LIFE CLEANSEA

Marine mammals, seabirds, turtles, fish and invertebrates have all been reported to ingest marine debris, especially plastic. Entanglement in marine litter has been reported for at least 20 pinniped species, at least 14 cetaceans, all seven species of marine turtles and more than 56 species of seabirds. The overall aim of the LIFE CleanSea project[8] is to support the application of the Marine Strategy Framework Directive (MSFD) and EU biodiversity policy relating to marine litter, with the objectives:

- To increase awareness of marine litter, empowering citizens to become part of the solution;
- To remove existing litter, including lost fishing gear, and prevent further littering;
- To promote "fishing for litter" initiatives and to train fishing industry professionals in responsible practices; and
- To provide guidelines for the management of marine litter, increase exchange of knowledge and the uptake of best practices and assist authorities in achieving a Good Environmental Status of the sea, as required under the MSFD.

4.1.7. LIFE SOUPLESS (Sustainable riverine PLastic removal and management)

One solution to prevent plastic waste from building up in oceans is to catch it in the rivers that are considered as the main pathway for transport of land-based plastic waste to the oceans. The LIFE SOUPLESS project[9] (“Sustainable riverine PLastic removal and management”) aims to demonstrate three new systems for riverine plastic removal and an innovative software tool to predict movement and accumulation hotspots of plastic litter in rivers. It will then demonstrate new water treatment techniques for removing micro-plastic from the rivers. LIFE SOUPLESS will then deploy innovative plastic recovery techniques at these pollution hotspots in an effort to maximise the efficiency of clean-up campaigns. One technique uses a helical coil that channels currents to guide litter towards a filtering barrier, a collection facility and a waste monitoring system. Another uses a device that blows a constant curtain of bubbles underwater, hindering the passage of litter and contaminants. Using

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[8] http://www.parcoasinara.org/it/contenuti/articoli/dettagli/521/
[9] https://webgate.ec.europa.eu/life/publicWebsite/index.cfm?fuseaction=search.dspPage&n_proj_id=6745

an additional pump and filtering module, project partners expect the bubbles to also halt the spread of microplastics in water bodies and help remove them from natural ecosystems.

4.1.8. WES (Water and Environment Support)

Started in 2019 with a 7.9 million budget, the WES project[10] ("Water and Environment Support in the ENI Southern Neighbourhood Region") will end in 2023. WES covers various regions such as Algeria, Egypt, Israel, Jordan, Lebanon, Libya, Morocco, Palestine, Syria, Tunisia. The aim of the project is to contribute to the reduction and prevention of the pollution in the Mediterranean region, to limit marine litter, to support the shift to a sustainable consumption and production model, to implement integrated management of water resources and to strengthen efficient use of water in countries highly vulnerable to the Climate Change. The project will build on the results of the initiative «Horizon 2020 for a cleaner Mediterranean».

4.2. COMPANIES DEALING WITH PLASTIC THERMOCHEMICAL TREATMENT

Caught in a fierce race to become more sustainable, oil majors, start-ups and downstream producers are navigating regulations, funding models, and technological developments to stand out as industry leaders in an emerging market, that of bio or alternative oil. Consumer demand for more sustainable gasoline and plastics will be key to future growth. Several companies are trying to satisfy this demand, such as Yamaha. Working in conjunction with Nexus Fuels of Atlanta, which is an end-to-end recycling business, and Tommy Nobis Enterprises, of Marietta, Yamaha Rightwaters[11] aims to return 10,000 pounds of polyethylene and polypropylene sheet plastics back into their base materials before the end of the calendar year. Yamaha Rightwaters is a national sustainability program that encompasses all of Yamaha Marine's conservation and water quality efforts. Program initiatives include habitat restoration, support for scientific research, mitigation of invasive species, the reduction of marine debris and environmental stewardship education. The fact that such a big and well-known company is focused on plastic recycling could help to increase people's awareness about the problem.

Another important company facing plastic waste thermochemical treatment is Resynergi[12], which is turning tons of floating plastic debris salvaged from the ocean into fuel as part of a program launched

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[10] https://www.wes-med.eu
[11] https://yamahaoutboards.com/en-us/utility/sustainability
[12] https://resynergi.com/news/fhcd6ut0hnujsaj46ag0ml2t7s447a

by Ocean Voyages Institute of Sausalito in collaboration with ByFusion Global Inc. of Los Angeles. The plastic is quickly heated using microwave energy and converted to gas. Gases are then condensed into liquid hydrocarbons such as usable oil, fuels and other products that can be reused to make new pure plastics. This process converts a ton of plastic waste into 200 gallons of fuel. This all-electric system is powered using 480 volts, does not include burning or incineration, and produces no smoke or other volatile organic compounds. Resynergi is also completing the design of a large capacity system capable of processing up to five-tons of waste plastic per day. The fuels produced could even be used to power the vehicles collecting the trash, to generate electricity or can be converted back into new usable plastics. A similar solution has been proposed by swiss record-breaking sailor and adventurer Yvan Bourgnon, who has unveiled a multi-functional research catamaran called "Manta"[13] that aims to de-litter the oceans. Thanks to a combination of four complementary collection devices, the Manta can capture floating macro litter as small as ten millimetres and as deep as one meter. The waste collected in collection carpets is lifted out of the water by inclined conveyor belts with a suction system and goes onto conveyor belts, where it is separated manually. Metals, glass, and aluminium waste are separated out, placed in two lifting containers of 40 tons each and later recycled locally on land. Organic waste ends up back in the sea. Plastic waste passes through a shredder and can be pressed into pellets. However, Frederic Silvert, a specialist in the Manta technology, points out that the material must be dry and - unlike household waste - desalinated and dechlorinated before further processing. Then a plant converts the collected, sorted plastic that can no longer be recycled into electricity. The plastic is then converted to syngas through pyrolysis, which enters a combustion chamber, and the resulting steam powers a generator and eventually a turbine that supplies electricity to all onboard equipment. The plan is to achieve energy autonomy of 50 to 55 percent. With this technology, the Manta will have a recycling capacity of 3.5 tons per hour in the future. Annually, this is expected to add up to 5,000 to 10,000 metric tons of plastic to be fished off at those locations that are considered the main sources of waste into the oceans. The catamaran is still in the planning stage, and according to Bourgnon, only a third of the costs have been covered. Nevertheless, he sees his project as the start of "a new generation of ships" with a different CO_2 footprint. He expects the current eco-catamaran to reduce CO_2 emissions by 75%.

Another important example of company involved in plastic recycling through pyrolysis is LyondellBasell, one of the largest companies in the world in the plastics, chemicals and refining sectors, which in September 2020 announced the launch of the MoReTec molecular recycling plant at the Ferrara

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[13] https://www.theseacleaners.org

site[14]. The new plastic materials created with this technology can be used for product sectors bound by strict regulatory requirements, such as food packaging and medical devices for the medical sector. The MoReTec technology development started in July 2018, when the company announced an active collaboration with the Karlsruhe Institute of Technology in Germany. In October 2019, the company announced the construction of a pilot plant in Ferrara. Today, thanks to the research and development of job groups in Germany, Italy and the United States, the company is actively working to explore potential commercial-scale applications. The pilot plant is capable of processing between 5 and 10 kg per hour of household plastic waste and it aims at understanding the interaction between the various types of waste in the molecular recycling process, to test different catalysts and to confirm the process temperature and the time required to decompose the plastic waste into molecules.

On February 2020, Eni Versalis launched the Hoop® recycling project[15], focused on the development of new technologies for the chemical recycling of plastics. The aim of the project is to develop innovative technologies for the recycling of plastics that cannot be recycled mechanically. Pilot-level testing and also the design of the demonstration plant with a capacity of 6,000 tonnes per year at the Mantova's site have already been completed. Now their aim is to scale up the process at a capacity of 30,000 tons per year.

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[14] https://www.lyondellbasell.com/en/news-events/corporate--financial-news/lyondellbasell-successfully-starts-up-new-pilot-molecular-recycling-facility/

[15] https://www.eni.com/en-IT/circular-economy/circle-plastics.html

5. INDUSTRIAL PYROLYSIS PLANTS

There are 87 active pyrolysis plants worldwide: 18 of them are industrially scaled up, which means they can treat at least 10,000 tons of waste per year, and 19 of them are at the demonstration step (10-1000 ton/year). Plants with a capacity less than 100 tons per year, which are those used in research centres, are called "paper plant". The most important companies dealing with plastic pyrolysis, or manufacturing pyrolysis plants, are listed in Table 5.

Company name	**Capacity (tons/y)**	**Input** (*)	**Oil output (%)**	**Website**
APC Pyrocrat systems	900÷30000	Plasmix	50 – 90	www.pyrocratsystems.com
Agilyx	3000	Polystyrene	50 – 90	www.agilyx.com
Alterra Energy	18000	Plasmix	50 – 90	www.alterraenergy.wpcom-staging.com
Anhui Orsun research technology	3000÷150000	PP, PE, PS	50 – 90	www.oursunchina.com
APChemi	23000	Plasmix	50 – 90	www.pyrolysisplant.com
Beston Machinery Co.	1800÷7000	PP, PE, PS	90 – 95	www.bestonmachinery.com
Doing Holdings	30000	Plasmix	45 – 52	www.doinggroup.com
Klean Industries Inc.	15000	Plasmix	N.A.	www.kleanindustries.com
Mingjie Group	6000÷7500	Plasmix	45 – 52	www.mingjiegroup.com
Nexus fuels LLC	15000	PP, PE, PS	Variable	www.nexusfuels.com/
Niutech energy LTD	7500÷15000	Plasmix	43 – 48	http://en.niutech.com/
TT Group recycling technology	15000	Plasmix	34 – 45	https://ttgroupworld.com/en/
Plastic2Oil	7500÷15000	Plasmix	84 – 88	www.plastic2oil.com

(*) as declared by constructor

Table 5. Summary of commercially available pyrolysis plants worldwide

The market role of these companies is mainly that of plant manufacturer, except for Agilyx, which is involved in styrene monomer production starting from polystyrene, and Nexus fuels and Plastic2Oil, which are fuel producers. Both batch and continuous plants are available. The type of reactor has an important impact in the mixing of treated plastics, use of catalysts, residence time, heat transfer and efficiency of the reaction towards achieving the final desired product (Anuar Sharuddin et al. 2016). Continuous plants are financially advantageous for industries converting large amounts of plastic waste, mainly because continuous processes help to save time. Labour costs are reduced as well: once the system is up and running, not much needs to be done aside from general supervision of the machinery (Goršek and Glavič 1997). The main advantage of a batch reactor, a closed system with no inflow and outflow of reactants and products while the reaction is running, is that high conversions can be achieved by leaving the reactant in the reactor for an extended period. On the other side, however, the disadvantages are the variability of product from batch to batch, high labour costs, and the difficulty of large-scale production. The composition of the output depends on the type of feedstock processed: for example, a large quantity of tyres in the input leads to the production of more coke. On the other side, polypropylene, polyethylene, and polystyrene treatment has shown to mainly favour oil production. Oil yield can be improved in two ways: using a catalyst or pre-treating the feedstock. An economic evaluation of the two cases is necessary to choose the most profitable solution. Post-treatment devices are used as well, to obtain a clean fuel and to avoid hazardous emissions. Many of the companies listed above are CE certified (Pyrocrat, APChemi and Doing) and they have plants running in EU countries. In this scenario, our aim is to scale up a pilot plant, the i-Cycle® technology (described in the next section), able to perform a pyrolysis process without particulate and hazardous emissions production, thanks to the presence of a stirred sphere reactor with three inner screw conveyers installed for the decomposition of polyhalogenated aromatic hydrocarbons in pyrolysis vapors. The scope is to obtain a simple, clean, and profitable process, able to reach high feedstock conversion, > 95%, and high oil yields, ≥ 90%. A heat exchanging system, which allows achieving superior heating rates to avoid PXDDF formations, is also part of the pilot plant.

6. CONCLUSIONS

Plastic waste production and disposal is a challenging problem, and a solution needs to be found as soon as possible to mitigate the environmental consequences of its wrong management. Nowadays several EU founded projects are facing this problem. Each project, as it has been shown in the previous sections, is focused on specific areas: awareness increase, modelling of plastic waste distribution and transportation, development of new technologies to collect plastic waste and microplastics, and development of new processes for plastic waste conversion into valuable products. Obviously, the results already obtained by previous projects and those that will be obtained from still active projects are all indispensable for a complete analysis of the phenomenon and for the development of industrial scale technologies. Some companies, like LyondellBasel and Yamaha, are already trying to scale up pyrolysis pilot plants which have been shown to give promising results in terms of yield and efficiency.

In the MARLESS project, the aim of the University of Bologna in Ravenna is to characterize plastic waste coming from Adriatic Sea and the oil produced after pyrolysis at Fraunhofer-Umsicht institute, in order to improve and refine pyrolysis conditions with the aim to obtain an oil with the same characteristics as those of diesel fuel. The i-Cycle® continuous pilot plant (Rieger et al. 2021), shown in Figure 3 with a capacity of 70 kg/h, will be used for the pyrolysis of the materials.

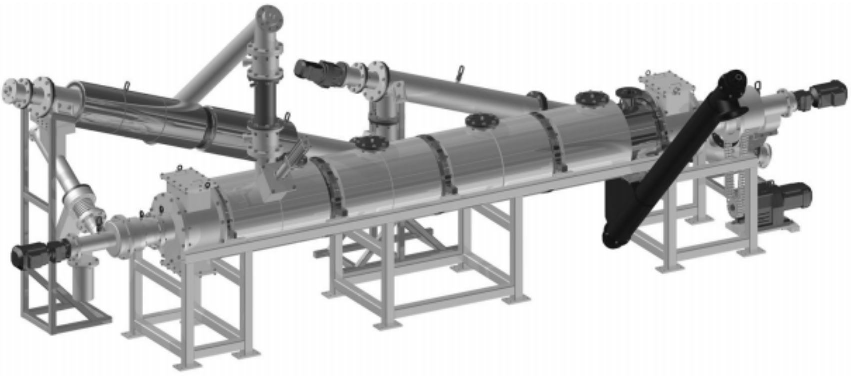

Figure 3. i-Cycle® pilot plant

The reactor of the pilot plant shows an externally and internally heated screw conveyer with a length of approximately 3.9 metres and an inner diameter of 0.5 metres. The feedstock will be characterized through elemental and thermogravimetric analysis. Other characteristics, such as water content and calorific value, will be determined as well. The pyrolysis oil will be analysed through gas chromatography-mass spectrometry. The aim is to develop a process without harmful emissions, which is environmentally friendly and economically advantageous.

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