Analysis of the use of recycled plastics

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Mechanical Engineering

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Abstract

There are significant environmental and public health concerns with the overuse and prolonged accumulation of solid plastic waste. Efforts to tackle this issue can rely on Circular Economy theories and strategies. For this reason, this work aimed to review literature not only on the environmental issue caused by solid plastic waste, but also on the theoretical background of Circular Economy and its applications. Based on Circular Economy strategies, a simple methodology to perform mechanical characterization of unknown recovered ocean plastic was then developed. Widespread industrial use of mechanically recycled plastics with unknown composition would not only present a sustainable source of material, but also help mitigate the pollution issue.

The methodology developed consisted of mechanically characterization specimens made of different mixtures of recovered ocean plastic waste, and their virgin counterparts, through tensile tests. Although the manufacturing of the test specimens, by means of a heat press, hindered the results that could be obtained, it also highlighted the importance of using a high-standard manufacturing process, such as extrusion. An appropriate manufacturing process will allow for detail in comparing virgin and recycled materials and for more reliability in increasing amounts of recycled percentages in plastic products (without significant loss of the materials' or parts' properties).

It was still possible to identify that even recovered materials of poor quality, which have been extensively degraded by exposure to elements in the ocean, present similar mechanical behavior of their virgin counterparts and can demonstrate reasonably good properties. The level of quality found in the recovered materials and the mixtures observed make them suitable for widespread use in industrial applications and the fast-moving consumer goods market, where plastic packaging is widely used.

In the future, appropriate manufacturing and virgin materials should be used to replicate different-percentage mixtures and evaluate their mechanical properties and how well they compare to the virgin counterparts.

Keywords: solid plastic waste, circular economy, mechanical recycling, tensile testing

Resumo

A acumulação de desperdício sólido de plásticos gera preocupações relativas ao meio ambiente e à saúde pública. Este trabalho produziu uma revisão bibliográfica que contribui para a caracterização das causas e dos impactos desta acumulação, particularmente em ambientes marinhos. A revisão incide também na teoria de Economia Circular, tendo sido identificadas estratégias que podem contribuir para a resolução do problema. Uma estratégia é a reciclagem mecânica de plástico recuperado do oceano. A utilização generalizada de plásticos mecanicamente reciclados representaria uma fonte sustentável de material e mitigaria o problema da poluição. No entanto, a sua adoção é dificultada pela heterogeneidade do material recolhido, que leva a incertezas acerca das propriedades mecânicas do material reciclado.

Foi desenvolvida uma metodologia simples para caracterizar mecanicamente plástico reciclado com origem em desperdício sólido de plástico recuperado do oceano. A metodologia desenvolvida consiste na realização de ensaios de tração para caracterizar mecanicamente provetes com composições diferentes de plásticos reciclados e dos seus equivalentes virgens. Recorrendo a esta metodologia, foi possível identificar que os provetes cujo material reciclado foi obtido a partir de materiais recuperados sujeitos a uma exposição prolongada aos elementos apresentaram um comportamento mecânico semelhante a provetes fabricados exclusivamente com materiais virgens.

O estudo efetuado apresenta algumas limitações devidas à utilização de um processo de fabrico pouco adequado, nomeadamente, a pressão a quente. Um processo de fabrico apropriado, como extrusão ou injeção, permitirá comparar os materiais virgens e reciclados com melhor fiabilidade e desta forma aumentar as percentagens de plásticos reciclados nos produtos (sem que exista uma perda significativa das propriedades dos materiais ou das peças).

O trabalho permitiu concluir que a aplicação de estratégias de Economia Circular pode contribuir para a mitigação do problema da acumulação de desperdício sólido de plásticos em oceanos, sendo possível a sua reciclagem mecânica e subsequente aplicação industrial dada a semelhança de comportamento mecânico exibido entre materiais reciclados e virgens.

Palavras Chave: desperdício solido de plástico, economia circular, reciclagem mecânica, ensaio de tração

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List of Acronyms

CE – Circular Economy

CNC - Computer Numerical Control

DIC – Digital Image Correlation

EU – European Union

HDPE – High-Density Polyethylene

LE – Linear Economy

LHPE – Low-Density Polyethylene

M1 – Type 1 manufacturing parameters

M2 – Type 2 manufacturing parameters

MFI – Melt Flow Index

NAFTA - North American Free Trade Agreement

OECD - Organisation for Economic Co-operation and Development

PP – Polypropylene

PE – Polyethylene

rHDPE – Recycled High-Density Polyethylene

rPP – Recycled Polypropylene

SPW – Solid Plastic Waste

vHDPE – Virgin High-Density Polyethylene

vPP – Virgin Polypropylene

List of Symbols

Latin Symbols

A – Nominal Strain at Brake

A_0 – Initial Area

A_i – Instantaneous Area

E – Modulus of Elasticity

e – Nominal Strain

F – Load

F_i – Instantaneous Force

l_0 – Initial Gauge Length

l_1 – Displacement of Point 1

l_2 – Displacement of Point 2

l_i – Instantaneous Displacement

Δl – Variation of the Gauge Length

S – True Stress

S_Y – Nominal Yield Strength

S_{ts} – Nominal Tensile Strength

t – Time

t_0 – Initial Thickness

v_0 – Initial Volume

v_i – Instantaneous Volume

w_0 – Initial Width

Greek Symbols

ε – true strain

σ – true stress

σ_Y – true yield strength

σ_{ts} – true tensile strength

1. Introduction

1.1 Motivation

Plastic is the most desirable man-made material not only due to its physical properties, (such as low weight and durability) and low manufacturing costs, but also due to its versatility in both design and manufacturing (Leal Filho et al., 2019), as well as excellent food protection capabilities (Eriksen et al., 2019).

Plastic production has increased almost tenfold, since large-scale production began around 1950. This happened mainly due to the replacement of reusable containers for single-use packaging (Geyer et al., 2017), which currently makes up for almost 40 percent of European demand by application (*PlasticsEurope*, 2019a). This trend is expected to continue, with predictions of a two-fold increase in plastic production over the next twenty-years (World Economic Forum, 2016).

Manufacturing of plastic products, which are in their majority non-biodegradable, leads to the accumulation of mismanaged plastic waste (Ritchie & Roser, 2018). This issue is especially relevant for ocean plastic waste as not only are there few ways of effectively monitoring this problem (Barnes et al., 2009), but also due to the extensive degradation of plastic exposed to the elements (Welden & Cowie, 2017). This degradation causes significant environmental impacts, including but not exclusively, for public health (Thompson et al., 2009). In its largest scale, plastic pollution affects the most occurring photosynthetic marine bacteria (*Prochlorococcus*), reducing its ability to produce oxygen. This issue is especially relevant considering this type of bacteria is believed to be responsible for up to 10% of global oxygen production (Tetu et al., 2019).

Figure 1.1 shows the route taken by plastic waste to the oceans from the stage of primary production until it reaches the ocean.

Figure 1.1 The pathway by which plastic enters the world's oceans (Ritchie & Roser, 2018)

Although it may seem like a small amount of plastic is leaking to the ocean, the figures are shown in an annual basis. Considering widespread use of food plastic packaging started in 1950 (Risch, 2009), the total plastic amount in the oceans is estimated to be over 500 million tonnes.

Mismanaged plastic waste ends up in the ocean due to different reasons. Not only is legislation more lenient and harder to enforce for disposal of plastic residues in water streams, but also due to leaking of nano and microplastics in everyday activities: shed of polymeric fibers by washing textiles, use of personal care products with microbeads (plastic particles with less than one millimeter in their largest dimension) and use of cleaning scrubbers (Browne et al., 2011; Eerkes-Medrano et al., 2015).

In a way to turn ocean plastic waste into a valuable resource stream and decrease the use of natural reserves of materials, Circular Economy (CE) presents several strategies. One of them is mechanical recycling. Finding mechanical properties for unknown source recovered polymers is a starting point to define appropriate mixture amounts of virgin polymers required and subsequent suitable applications. These applications should maximize the lifetime of the manufactured products to prevent them to re-enter pollution streams. Examples of possible industries that can benefit from the use of recycled plastics are building and construction, furniture and automotive.

1.2 Objectives

The main objective of this dissertation is to develop a simple methodology to perform mechanical characterization of mixtures of different percentages of unknown recovered ocean plastics and their virgin counterparts.

This methodology will be based on:

1) A literature review on Circular Economy (CE) for ocean plastic valorization
2) Mechanical characterization of plastics through tensile testing

Initially this project aimed to test the ocean plastic material in order to produce products, which would valorize the plastic waste, however due to the circumstances surrounding the COVID-19 pandemic, most of the initial plans had to be redesigned. Therefore, a deeper literature review had been added as a main goal of this dissertation, allowing to identify and analyze the possible processes that can be used to valorize ocean plastic waste to final products in a CE perspective. Mechanical characterization of the recovered materials will make their widespread industrial use possible because their properties are known and can be adjusted to different applications. Figure 1.2 shows the interaction and overlap of CE theory, environmental concerns about solid plastic waste (SPW) and mechanical characterization of materials.

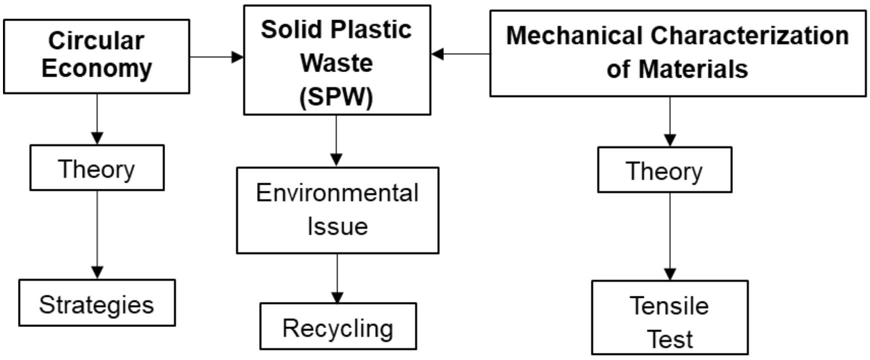


Figure 1.2. Work structure considering CE strategies and mechanical characterization to mitigate SPW's environmental issue

1.3 Structure of dissertation

This thesis is organized in five chapters, including this introduction.

Chapter 2 presents the State of the Art starts by describing plastics, its markets and manufacturing processes, and the environmental and public health issues that arise from its use and accumulation. It then describes the environmental issue and potential consequences surrounding marine solid plastic waste. A characterization of this waste as well as its supposed location is also given. Afterwards, definitions and strategies of CE are explored and the opportunity for mechanical recycling of plastic waste is explained. Finally, an in-depth analysis of mechanical recycling is done, and the variability in properties in recovered plastics shown. The issue of variability then explains the need for a standard methodology for mechanical characterization. Afterwards, the way mechanical characterization is done is explained, and finally, examples of characterization done with mixtures of recovered and virgin materials are shown.

Chapter 3 presents the proposed methodology, the materials used, and the groups of specimens tested. The manufacturing method, as well as the processing parameters used during manufacturing, are also shown.

Chapter 4 shows the results obtained for the tests performed and discusses the validity of the results, the limitations encountered during the processing of the materials, and the contribution and outcomes of the research done.

Finally, in Chapter 5 a conclusion is given, and future work is discussed and proposed.

2. State of the Art

Since the objective of this work is to develop a simple methodology to test recycled ocean plastic, this chapter focuses on the challenges and opportunities relating to this material and its processing. Section 2.1 begins by presenting the main characteristics of plastics and their markets, both for virgin and recycled materials, as well as manufacturing processes. Additionally, this section presents the environmental issues and impact caused by solid plastic waste (SPW). To mitigate this issue, Circular Economy (CE) is explored in Section 2.2, both in its definition and principles, and in its strategies as well. Section 2.3 presents mechanical recycling of plastics as a CE strategy and explores this process. In Section 2.4 one of the main barriers for mechanical is presented: thee variability of recycled plastics' mechanical properties. To counter this issue, mechanical characterization methods are presented in Section 2.5. Finally, in Section 2.6, a review of literature of the characterization of mixtures of recycled and virgin plastics is presented.

2.1. Plastics

2.1.1. Materials

"Plastics" describe organic materials produced from natural, organic materials. These can be cellulose, salt or fossil fuels. Most plastics are derived from fossil fuels. The production of plastic from crude oil is done from a product of distillation in oil refineries, or fractions. Fractions are mixtures of hydrocarbon chains, and naphtha is the main fraction from which plastics are produced. Figure 2.1 shows the flowsheet of the steam cracking process. Common polymers like ethylene and propylene are end products from the processing of fractions – at the hydrocarbon fractionation and hydrogenation stage - from the distillation of crude oil – which is the initial stage of gas or liquid feed.

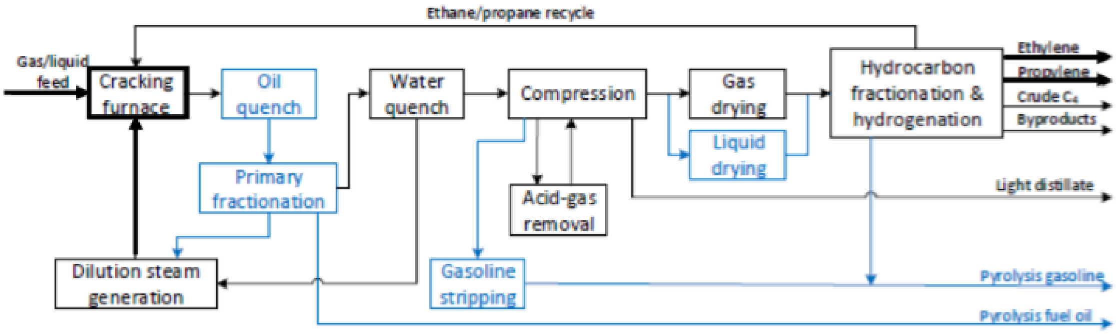
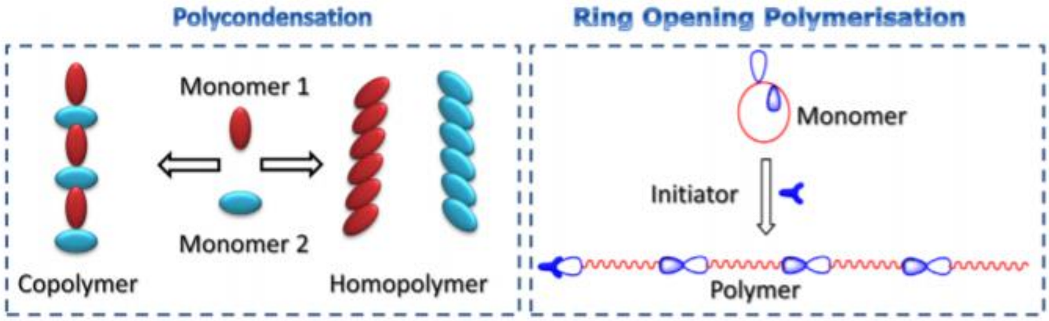


Figure 2.1. Simplified flowsheet of the steam cracking process (Marcos, 2016)

The two main processes used to produce plastics are polymerization and polycondensation. In both, the goal is to link monomers – single molecules that are the functional unit of polymers – and turn them into polymers – long chains with improved mechanical properties. In polymerization a reactor is used to bond monomers such as ethylene and propylene, usually by adding catalysts, and form long polymer chains. Polycondensation is a condensation reaction of monomers with two functional groups that form a polymer (polycondensate). Figure 2.2 shows monomers as the building blocks for polymers, both the polycondensation and polymerization processes, and the way the single functional units turn into long polymer chains.



(a) (b)

Figure 2.2. (a) Schematics of polycondensation (b) ring-opening polymerization reactions (Bansal et al., 2016)

The extent of combinations and properties that can be achieved with both plastics and their additives are the main reasons why they became so widely spread in such a short amount of time (Van Eygen et al., 2017). Depending on the monomers used, plastics can have different characteristics. In general, they are widely used due to the following properties:

- Lightweight – relative low weight and good mechanical properties
- Water – protection – this is a very relevant benefit of plastics as it makes them the preferred materials for packaging applications where moisture or air must be kept out. One prominent example of these applications is food-protection packaging.
- Versatility – plastics can be made into a lot of different geometries and dimensions and keep their mechanical properties
- Low processing cost – comparing with the production of other engineered materials such as steel and cement, plastics can produce mass series of products with relatively low costs. They are also derived from products of existing oil refineries, making it a profitable side-industry

- Durability – because plastics have extremely good water and air-protection properties, they are extremely resilient to weathering and degradation in conditions most materials would not last. For this reason, they are very durable.

Comparing plastics to other engineering materials shows a trend for increase in consumption. Engineering materials derive from oil (polymers), ores (metals and ceramics), and biomass (timber and paper). Figure 2.3 shows the historical evolution for key engineering materials through time.

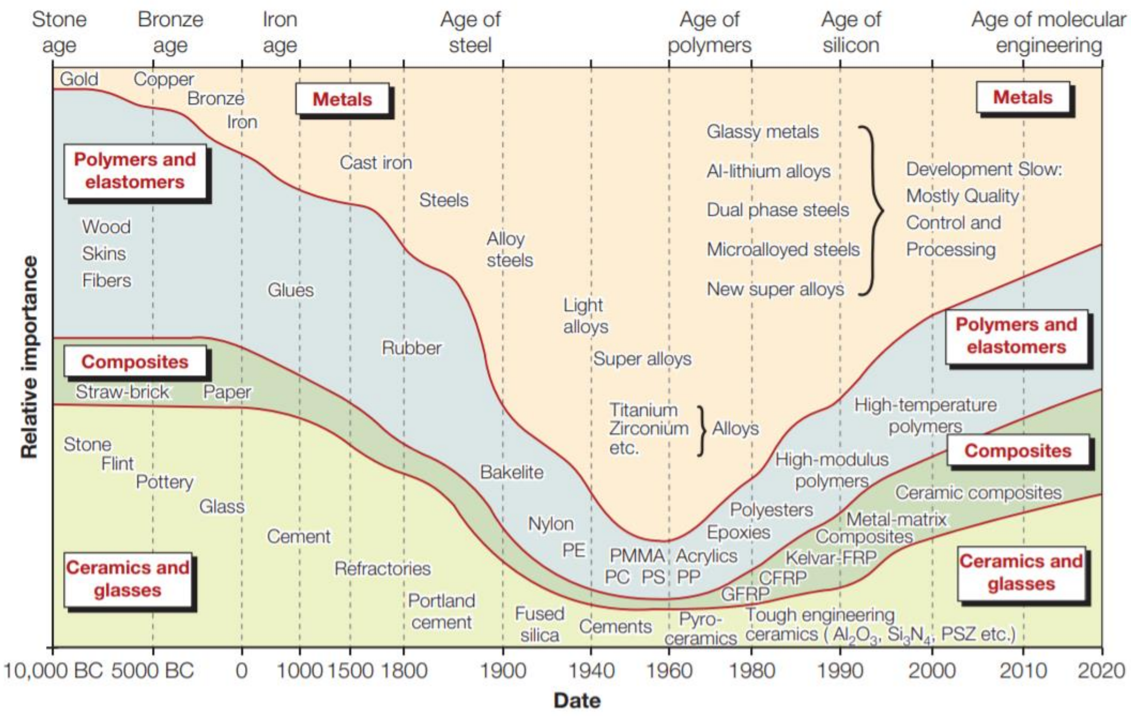


Figure 2.3. Evolution of materials' importance with time (Ashby, 2011)

Although not immediately apparent from Figure 2.3, plastics have shown the strongest demand growth since they were first produced, surpassed only by metals. Still, plastics pose an extra difficulty to replace because they are dominant in the packaging sector, having replaced a lot of the more “traditional” materials like aluminum and paper. Additionally, the issue with plastics is that they mainly derive from oil and ores, considered to be non-renewable. This means plastics will inevitably reach a point in which their cost will exceed their utility (Allwood et al., 2011). Lastly, it is worth noting that should relative importance for all materials stabilize, eventually reaching an equilibrium, there is a possibility of creating a closed-loop system. The only way to create closed-loop plastic systems though will be to move production almost entirely to recycling operations, (considering demand will still increase for plastics) (Allwood et al., 2011). A closed-loop system allows for a more sustainable management of natural reserves (and consequently of waste. This concept will be explained in the CE section of this chapter.

2.1.2. Market for virgin plastic

Considering European Union plastic market had a turnover of about 340 billion euro in 2015 (European Commission, 2018) and the market share for Europe was of about 20% in 2016 (OECD, 2018), as estimate for the global plastic market can be made at about 1700 billion euro. Figure 2.4 shows the global demand for plastic products by segment.

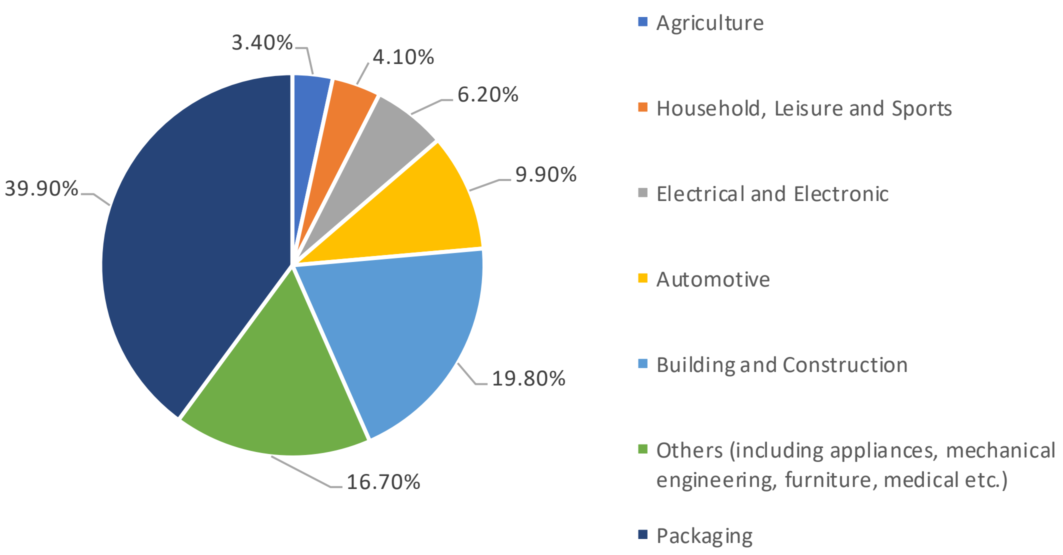
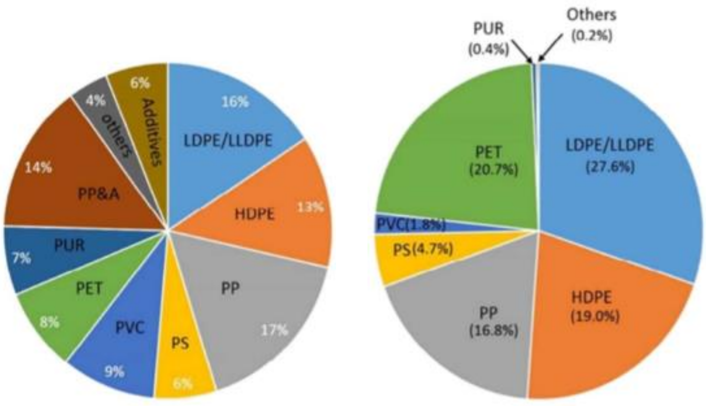


Figure 2.4. Global plastic demand by segment (PlasticsEurope, 2019b)

Packaging represents almost 40% of plastic products produced worldwide. Due to its nature, plastic packaging is often single use. While other applications also represent big amounts of plastic produced worldwide, packaging is very difficult to replace due to its properties and convenience. As will be explained in the CE section, plastic's short life span means this material is a stream of little value, since its "nutrients" are lost as soon as it is produced.

Since packaging is the most widespread application, Figure 2.5 shows the global production of different types of plastic produced and the ones used for packaging. As expected, common plastics used for packaging are the most common plastics produced. These include PP and LDPE/LLDPE and HDPE. All of these are thermoplastics, which, as mentioned before, can be melted, and reformed several times.



(a) (b)

Figure 2.5. (a) Types of plastics produced worldwide (b) plastic use in the packaging industry in the world in 2015 (Royal Society of Chemistry, 2020)

In Figure 2.6, the global plastic production by region/country is shown.

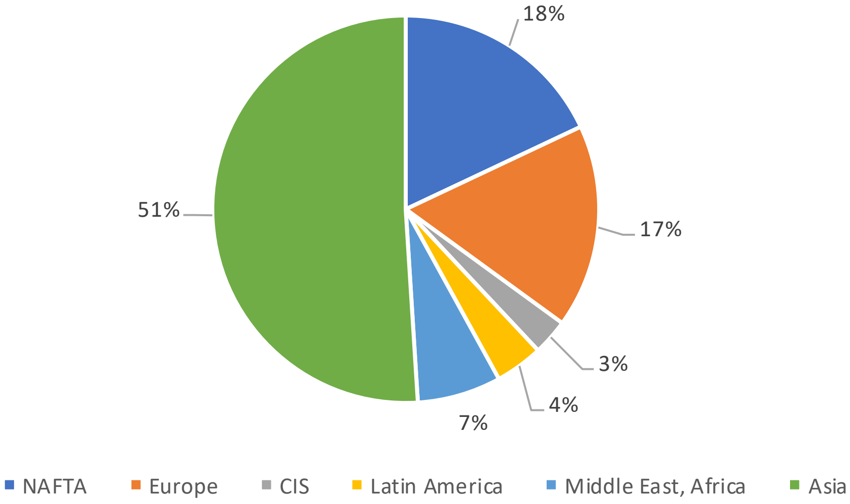


Figure 2.6. Global plastic production distribution by region in 2018 (PlasticsEurope, 2019b)

It is worth noting that that the North American Free Trade Agreement region (NAFTA), Europe and Asia are responsible for more than 85% of annual plastic production. In these countries, plastic waste is the largest amount by weight of non-biodegradable municipal solid waste (Wang & Nie, 2001). It can be concluded that countries producing and consuming the most plastic are dealing with the waste created by this material not only at an idustrial level but also at a consumer level. The environmental and public health issue of plastic waste will be further explored in the next section. To mitigate this issue, turning this waste into a feasible source of material itself could pose as viable solution.

2.1.3. Market for recycled plastic

The recycled plastics' supply chain is complex for two reasons: not only are there several pathways for plastics to go from point of "initial" manufacturing to where they become recycled materials, but also because of the great variety of polymers and combinations possible amongst them. Figure 2.7 shows an overview of the plastics industry structure.

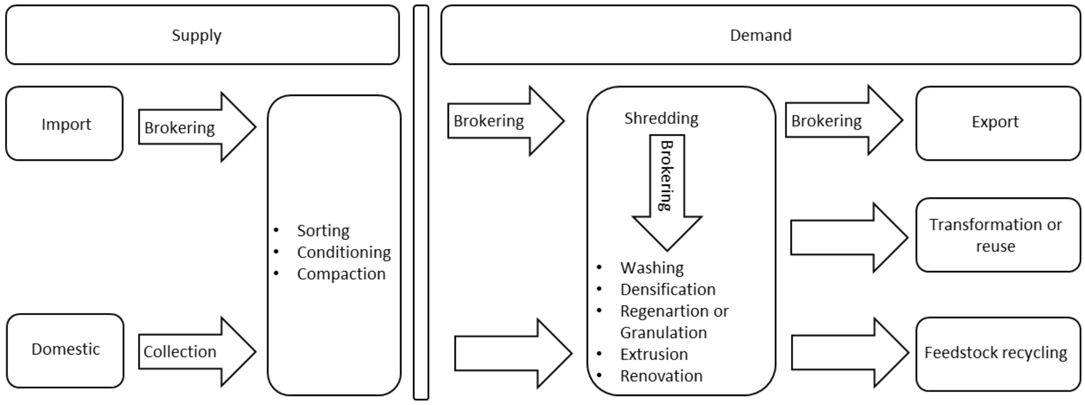


Figure 2.7. Simplified recycled plastics supply chain, adapted from OECD "Improving markets for recycled plastics"

Brokering refers to an entity that markets materials on behalf of a second party (for a commission of the revenue generated). The figure shows a similar infrastructure to the one for virgin plastics. The different operations shown can be carried out by a single party at different locations, in a single location, or by multiple parties in a singular or multiple location. These activities can also be carried out by commercial operators, governmental agencies, or non-governmental organizations. All these factors make for hard homogenization of the recycling process, and consequently, for low success rates. Demand for recycled plastics varies with many factors. The main ones are described in Table 2.1.

Table 2.1. Factors influencing demand for recycled plastics – adapted from (OECD, 2018)

Consumer demand	Clothing
	Replacement of metal and ceramic products such as construction materials and automotive parts
	Competing products (wood, paper, and reusable items)
Environmental policy	Producer responsibility legislation
	Corporate social responsibility agendas
	Public sector procurement policies favoring recycled content
Enabling technology	Extrusion and forming - enabling higher content of recycled material
	Product specification

As expected, consumer pressure lays a big role in demand for recycled plastics. Additionally, prices for recycled plastic can sway greatly according to policy and economic incentives for recycling industries or taxation for extraction and manufacturing of virgin material. Technology also plays a significant part in demand because as the processes are optimized, they become more profitable and the industry grows.

There are different stakeholders in the recycled plastics industry, namely governmental organizations, and private companies, both usually complying with regulations and legislation on waste management. Additionally, community-based organizations and non-governmental organizations can also assume roles in the recycled plastic industry.

For a better understanding on how the cost of recycled plastic compares to the one of virgin plastics, Figure 2.8. Market value for major polymers 2012-2015 presents the market value for major polymers for a period of 3 years starting in 2013.

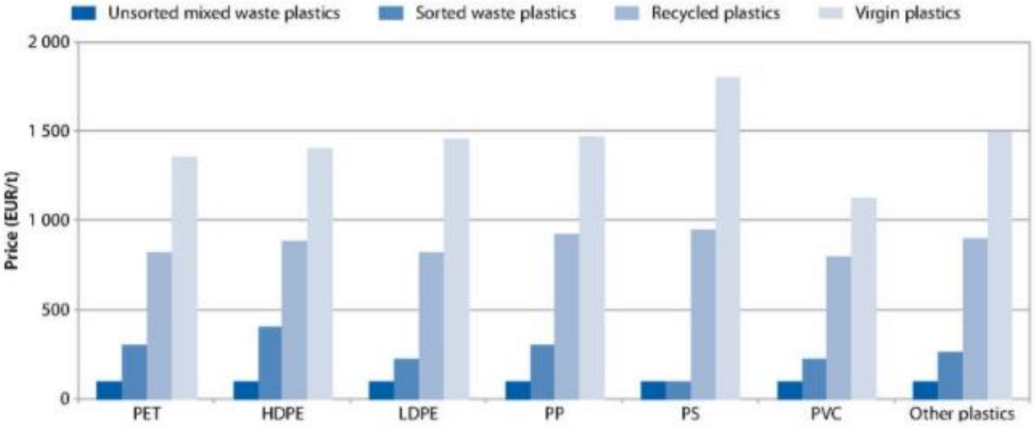


Figure 2.8. Market value for major polymers 2012-2015 (OECD, 2018)

It is worth noting that the Chinese plastic ban in 2017 has since disrupted the recycled plastic market, forcing developed countries to recycle and process plastic waste domestically and driving prices for recycled plastic to increase. It can be seen from the plot that the most recycled plastics are the ones most produced, which then leads to an assumption that technology is not hindering mechanical recycling rates.

Several factors can influence prices for both virgin and recovered plastics. These are summarized in Table 2.2.

Table 2.2. Main drivers for prices of virgin and recycled plastics (OECD, 2018)

Driver	Examples	Virgin plastic	Recycled plastic
Operational Costs	Grid energy Real estate Labor Processing		X
Oil price	Cyclical trends Trader behavior (OPEC influence) Supply and Demand Other fuel markets (e.g., shale oil)	X	
Cost of additives	Plasticisers/softeners Catalysts Flame retardants	X	X
Supply and demand (of plastic)	Stockpiling (artificial market inflation) Cost of substitutes (e.g., paper, wood)	X	X
Cotton price (PET only)	Clothing market supply and demand Cotton quality Cost of substitutes (e.g., wool) Stockpiling	X	X
Policy	Legislation mandating weight or proportion of plastics that must be recycled (creating an economy of scale)	X	X
Global supply chain networks	Westbound freight costs and backloads to Asia on empty ships		X
Technological capability	Processing Logistics		X

Some of these factors are expected to influence prices in a lot of industrial processes, such as most operational costs, global supply and demand and technological capabilities. But some aspects are specific to plastic markets. The production of plastic is a highly energy intensive process, so fluctuations of energy prices have a high impact on plastic trade prices. On the other hand, the most globally produced plastics derive from olefins, so oil prices are another factor that has a great impact on plastic prices. As mentioned before, policy and taxation can sway plastic prices (virgin and recycled), even if not accounting the other impact factors. Notably, price of substitutes for plastic also influences its price, especially in the textile market, where polyester can be replaced by cotton or wool. Other external factors like stockpiling and backloads or political instability can also manipulate the plastic market and influence prices.

Recycled plastics are usually competing in the same markets as virgin ones except for in the construction materials' industry where they have for long been an acceptable and widely used alternative to virgin polymers.

2.1.4. Manufacturing

Most of the manufacturing of plastic parts can be divided into extrusion, injection, and blow molding (Martins & Rodrigues, 2010) although more recent alternatives like computer numerical control (CNC) machining and 3D printing are now also possible (Groover, 2013). To choose the appropriate manufacturing process the following parameters should be taken into consideration:

- Type of polymer (and its properties, such as melt flow index (MFI), viscosity and melt temperature).
- Geometry of the part – simple geometries with little differences in thicknesses and radii do not require advanced processes, while more complicated ones need to be done with special molding, CNC machining or 3D printing.
- Volume/cost – some processes, like injection molding, require the machining of molds which can represent a high cost per part if the volume of the series is not big enough. For smaller series or prototypes, newer processes such as CNC machining or 3D printing can be more suitable.
- Lead time and manufacturing time – when having a small lead time, processes requiring molds or casts might not meet requirements because it takes time to produce these molds. On the other hand, machining time also varies from process to process so this can be a decisive factor.

Perhaps the most significant factor in choosing the manufacturing process is the ratio of volume to cost (Groover, 2013). Figure 2.9 shows which processes are more suitable depending on the volume of the series and cost objectives (*Guide to Manufacturing Processes for Plastics*, n.d.). For prototypes or small series, CNC machining, vacuum forming, polymer casting and 3D printing are the most appropriate process. For a small to medium volume of parts, rotational molding, extrusion, blow molding and injection molding can be chosen, depending on additional requirements or objectives. While most processes become more profitable as volume of series increases (meaning a lower cost per part), others reach a point where their cost and/or capacity no longer justifies their use. This would be the case for 3D printing, per example, where producing a series of 100,000 parts would take too long and be too costly.

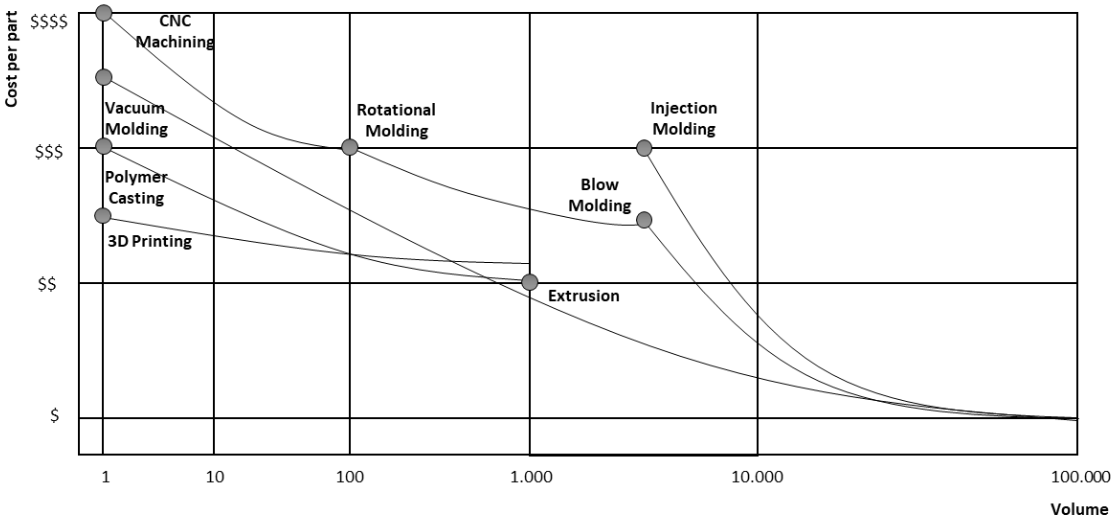


Figure 2.9. Plastic manufacturing processes compared regarding cost and series volume, adapted from (*Guide to Manufacturing Processes for Plastics*, n.d.)

Table 2.1 further explores different relevant aspects of plastic manufacturing processes and makes qualitative comparisons between processes' geometry capabilities and associated costs.

Table 2.3. Comparison of different aspects of plastic manufacturing processes - adapted from (*Guide to Manufacturing Processes for Plastics*, n.d.)

	3D Printing	CNC Machining	Polymer Casting	Rotational Molding	Vacuum Forming	Injection Molding	Extrusion	Blow Molding
Geometry complexity	●●●●●	●●●●○	●●●●○	●●○○○	●○○○○	●●●●○	●○○○○	●○○○○
Lead time	12 - 36 hours	24 hours – 1 week	24 hours – 1 week	4 – 6 weeks	4 – 6 weeks	8 - 10 weeks	2 – 4 weeks	4 – 6 weeks
Cycle time	< 1 hour to multiple hours (depending on part size and volume)	< 1 hour to multiple hours (depending on geometry and part size)	Minutes to multiple days (depending on curing time)	< 1 hour	Seconds to minutes (depending on equipment)	Seconds	Seconds (or continuous)	Seconds
Set up cost	$	$$	$	$$$	$ - $$$$	$$$$$	$$$	$$$$
Cost per part	$$$	$$$$	$$	$$	$ - $$$	$	$	$
Volume	1-1000 parts	1 – 5000 parts	1 – 1000 parts	100 – 5000 parts	Any volume of parts	+5000 parts	+1000 parts	+5000 parts

As expected, processes appropriate for big series produce parts with more simple geometries, and processes more suited to small series can produce more complicated shapes. Another significant point of Table 2.3. is the comparison of lead time amongst processes. As mentioned before, processes requiring molds or casts call for a significantly higher lead time in order to produce these parts. Processes better suited for prototypes like 3D printing can be set up very fast and adapt do design changes very quickly. All these processes will be further explained in this section.

The main plastic manufacturing processes are described in more detail in the next sections.

Polymer Casting

This process is very simple and one of the oldest. Figure 1.1 shows simple schematics for polymer casting and the part produced.

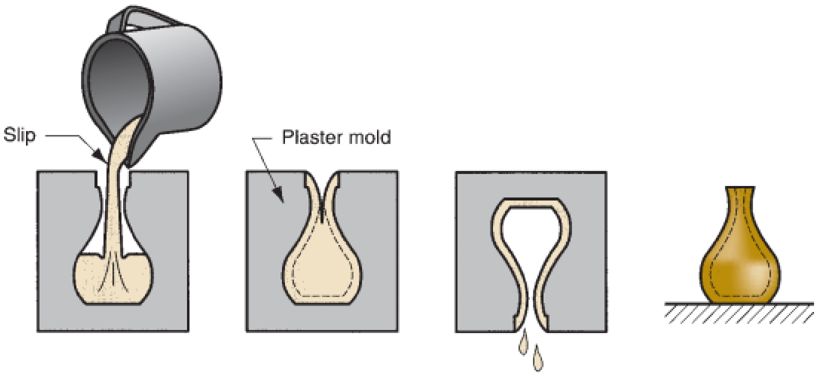
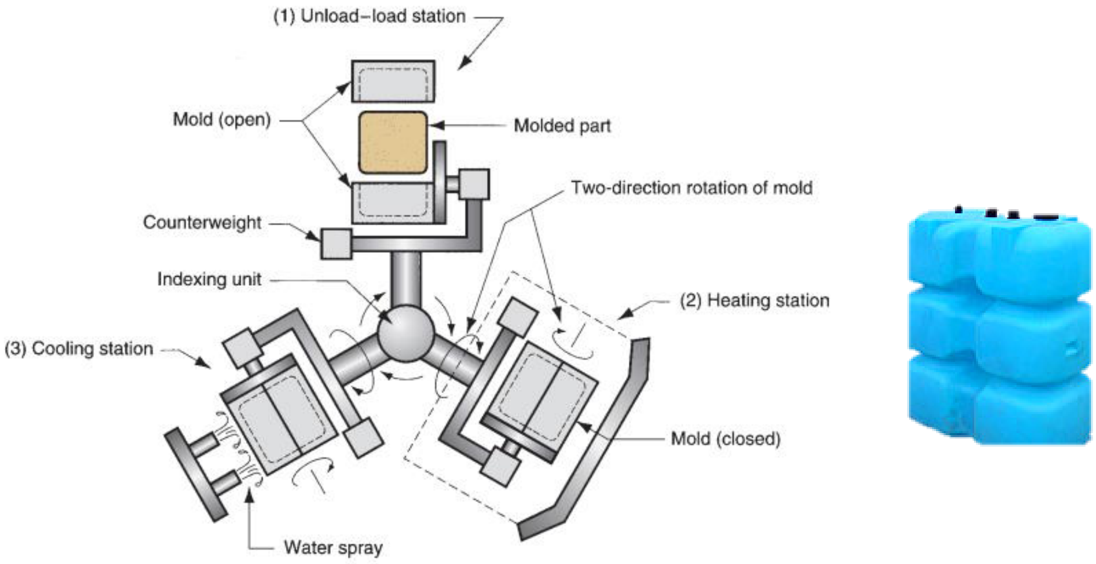


Figure 2.10. Polymer casting diagram and part (Groover, 2013)

The process consists of pouring reactive liquid synthetic resins in flexible molds made from latex or silicone rubber. The material then cures or hardens because it can be mixed with a curing agent, and/or is heated to accelerate its solidifying. The mold is the negative to the part's positive. This process involves some preparation of the mold with a release agent to facilitate demolding and can also be necessary to preheat it. This process is highly suited for detailed geometries and parts with fine features. Due to the molds' flexibility it is more advisable for solid parts. Polymer casting is a relatively inexpensive process, but it requires manual labor after the part is produced so it can have a high cost per part comparing to automated processes like injection.

Rotational Molding

This process is also called rotomolding. Figure 2.11 shows simple schematics of this process and a part produced.



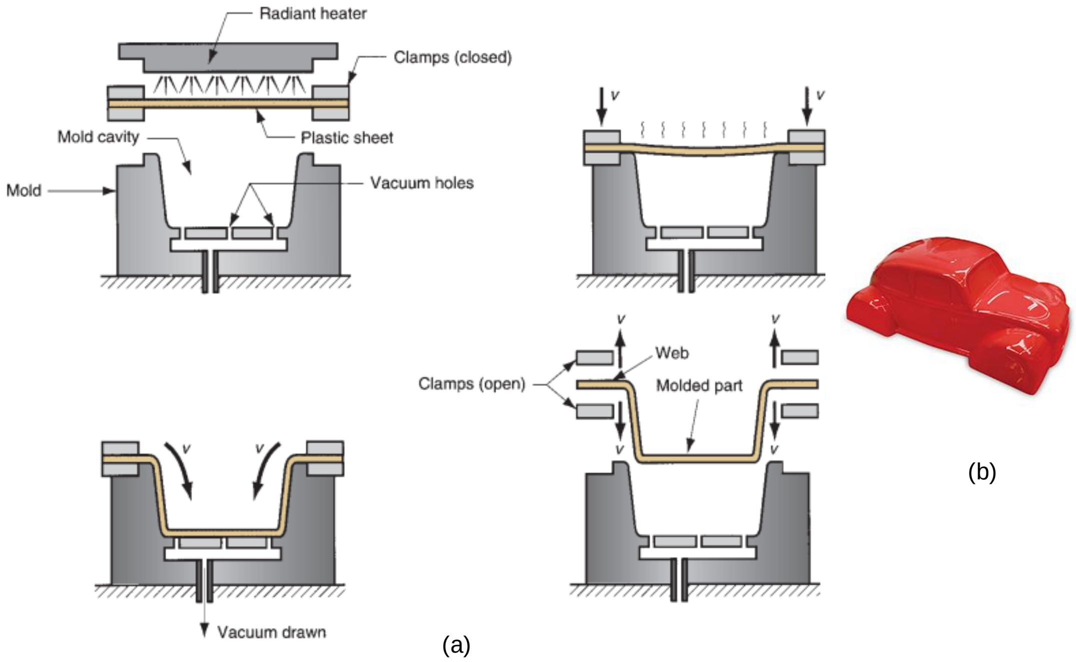
(a) (b)

Figure 2.11. (a) Rotational molding diagram (Groover, 2013) (b) container produced by this process (Licomplast, n.d.)

This part has a simple geometry, uniform wall thickness and it is hollow. Examples of parts possible to make are tanks, containers, helmets and canoe hulls. Powdered plastic is deposited in the mold cavity and is then rotated along two perpendicular axes while inside a heating station. The powdered thermoplastic melts and coats the cavity building the part and the rotation assures a uniform distribution of the material. Afterwards, the mold is cooled through convection or water in a cooling station, and finally the part is demolded. This process doesn't require expensive tooling as it relies on centrifugal force to fill the mold, as opposed to pressure like in most other processes. Because molds need to be heated and cooled, this process has long cycle times, making it unsuitable for high volume applications. Once set-up is done, cost per part is relatively low for this process, especially for large parts. Rotational molding is suited for simple, large and hollow parts, with loose tolerances.

Vacuum Thermoforming

Figure 2.12 shows simple schematics for this process (a) and a part being produced (b). In this process, a clamped plastic sheet is heated and formed using a mold. The mold can be below the plastic sheet, above, or the part can be produced using two molds, one on each side of the sheet. Once the plastic is stretched over the mold, a vacuum system removes the air between the sheet and the mold, forming the part.



(a)

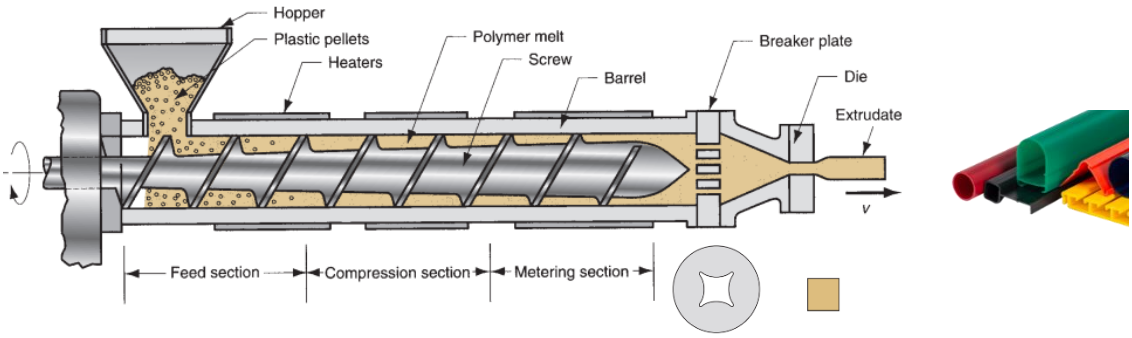
(b)

Figure 2.12. (a) Vacuum forming diagram (Groover, 2013) (b) part produced (Europlas, n.d.)

Produced parts need to cool before being completely set, either by room temperature convection, with a fan system or by spraying fluids. Some post processing trimming, and additional finishing might be required, either by hand or CNC machining. Molds used are made from wood or resins and are only suited for custom parts like prototypes or low volume series. The process has some limitations regarding geometry, and it is appropriate for thin-walled parts only.

Extrusion molding

Like injection molding, extrusion is one of the most used mass production process of plastic parts. Figure 2.13 (a) shows the schematics of this process. Solid material (pellets) are fed to the machine in the hopper and, much like injection molding, are then heated by heaters around the barrel. As a screw rotates, the molten plastics is forced through a die (Figure 2.13 (b)). The orifice in the die is the cross-section of the extrudate produced (Figure 2.13 (c)). The continuous profile is then spooled or cut.



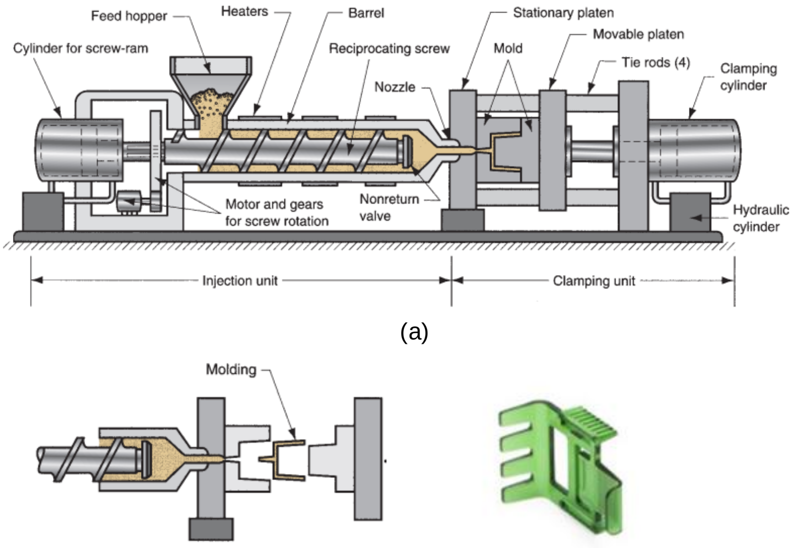
(a) (b) (c) (d)

Figure 2.13. (a) Simplified extrusion molding diagram (b) example of a die (c) extruded profile (Groover, 2013) (d) examples of profiles obtained by extrusion molding (Gabriel Trade, n.d.)

The die is the “mold” of this process and must be machined to fit the geometry of the final profile. Extrusion molding is not as expensive as injection molding, for example, because the dies are very simple and don’t require such tight tolerances. Like injection molding, this process offers short cycle times making it economic for high volume series. Figure 2.13 (d) shows examples of profiles obtained by extrusion molding. Possible geometries for profiles are somewhat limited, like sections in a T, I, L, U or other simple shapes like circles or polygons. This process is ideal to manufacture pipes and straws or buildable frames.

Injection Molding

Injection molding is the most used manufacturing process for mass production of plastic parts. It works by forcing molten thermoplastic under high pressure into a mold cavity, which is then closed by a hydraulic press. Figure 2.14 shows the schematics for an injection molding installation and an example of a part that can be produced through this process.



(a)

(b) (c)

Figure 2.14. (a) Simplified injection molding diagram (b) detail of the molding produced (Groover, 2013) (c) example of a part produced

Plastic pellets are fed to the machine in the feed hopper and are then pushed through the screw and heated with bands around the barrel. The heated plastic is then pushed through the nozzle into the mold cavity, where it solidifies. This process is used in mass production of parts because cycle times for parts are extremely small, ranging from 10-30 secs for small and medium parts.

Injection molding are able to produce very complex parts, and for these molds need to be manufactured with tight tolerances. Often, special care needs to be taken with demolding and extra channels or inserts need to be used. Because there are high temperatures and pressure involved (comparing to other processes), molds are machined in metals, such as hardened steel. Depending on the metal used in the molds – harder or softer – more or less parts can be produced from the same mold, respectively. All the requirements for the molds, as well as their cost, make this process suited only to high volume series with a big lead time (tooling can take months to be prepared at high costs). Once the set-up is prepared, this process can offer an extremely low cost per part because one mold can produce millions of quality parts extremely fast. Post processing of injected parts include removing sprues, runners and any potential flash from the part. This can often be done automatically as part of demolding. Most thermoplastics can be used in this process, and for thermosetting plastics, reaction injection molding (RIM) is the equivalent process. Figure 2.15 shows the schematics for RIM. With RIM, structural plastic foams are produced through chemical reactions between liquid plastics. These liquids are mixed in the mixing head at high pressures and are then injected into the mold cavity. In the mold cavity they react and polymerize, forming a foamed part shaped like the mold cavity.

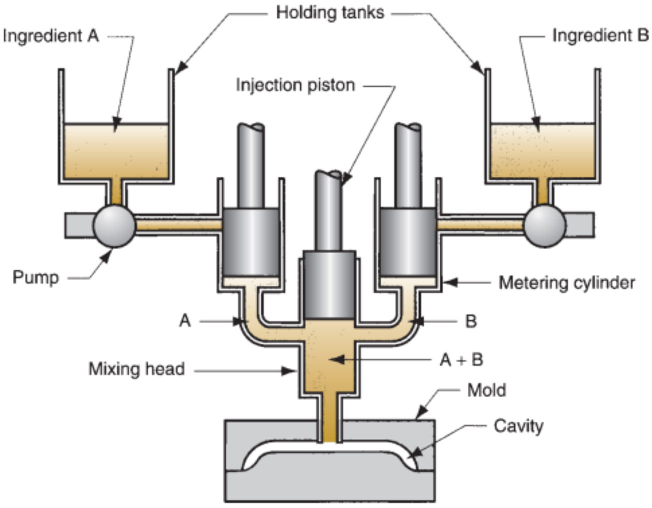


Figure 2.15. Simplified reaction injection molding diagram (Groover, 2013)

Blow molding (extrusion or injection)

Like rotational molding, blow molding is used to produce hollow shapes, preferably seamless. Figure 2.16 shows the schematics of this process for both extrusion and injection blow molding. In both processes, the final shape is created by effectively inflating an initial shape with air.

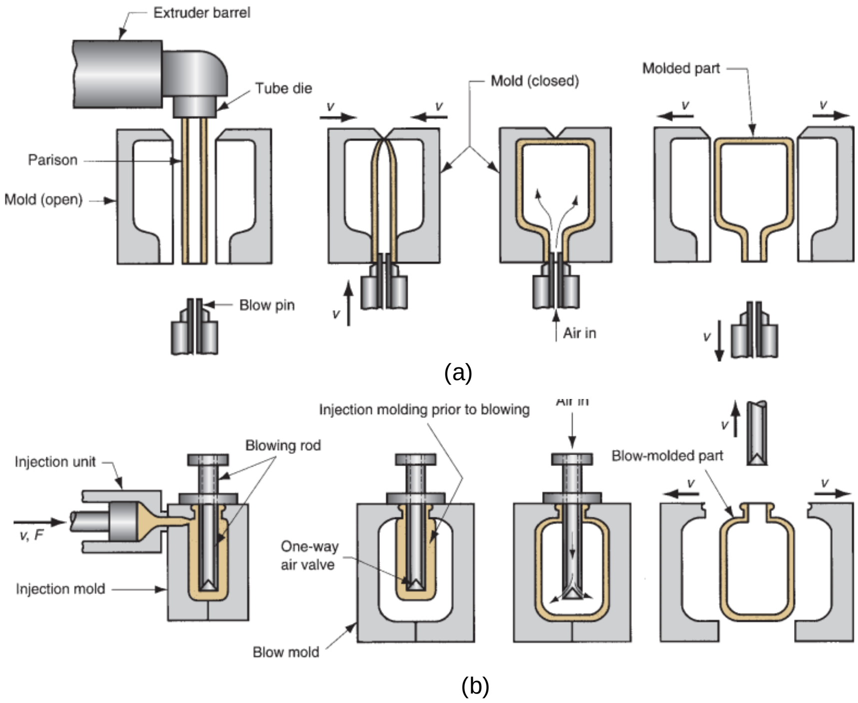


Figure 2.16. (a) Simplified extrusion blow molding diagram (b) injection blow molding (Groover, 2013)

This initial shape is called a parison for the extrusion process and a preform for injection. In extrusion blow molding, the parison is created by forcing melted plastic pellets through a die, while in injection molding, the molten plastic is forced by the screw into a mold, creating the preform. Both are then inflated and demolded. This process has a lower tooling costs than injection molding because it operates at much lower pressures. Like injection and extrusion molding, blow molding is a continuous process, making it economic for mass production of plastic parts.

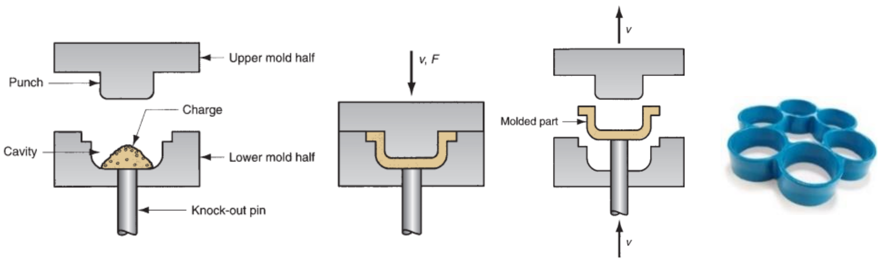
Figure Figure 2.17 shows perhaps one of the most well-known applications of this process: a plastic bottle and its mold.

Figure 2.17. A mold for a plastic bottle, a parison or preform and the final plastic bottle obtained through blow molding

With the final product, the mold used to produce it can be seen, as well as the parison or preform used. This process is suited for hollow and thin-walled products, with simple shapes.

Compression molding

This process was one of the first to be developed for the manufacturing of plastic and it is widely used for thermosets. Figure 2.18 shows simple schematics for compression molding.



(a)

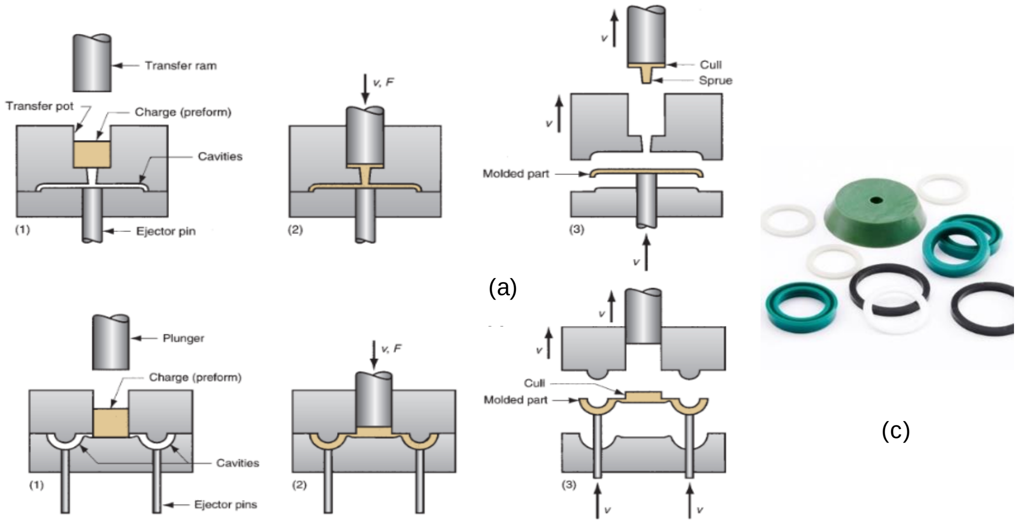
(b)

Figure 2.18. (a) Simplified diagram of compression molding (Groover, 2013) (b) part produced by this process (Plan Tech, n.d.)

And consists of loading an amount of plastic called the charge into the lower mold half, which has been heated in advance. The upper part of the mold is then forced into the lower one and a punch forces the charge to flow evenly to the cavity. The heat applied polymerizes and cures the plastic and a solidified part is then obtained once the two molds are opened.

Transfer molding

This process is similar to compression molding, but in this case, the charge is loaded to a chamber immediately ahead of the mold cavity. Figure Figure 2.19 presents the two variants of transfer molding: (a) pot transfer molding and (b) plunger transfer molding.



(a)

(c)

(b)

Figure 2.19. Simplified diagrams of transfer molding: (a) pot transfer molding (b) plunger transfer molding (Groover, 2013) (c) example of a parts produced by this process (Archer Advanced Rubber, n.d.)

The processes differ only in that in pot transfer molding the charge is insert by a channel directly into the cavity, whereas in plunger transfer molding the charge flows to lateral channels into the cavity.

Heat pressing

In this process plastic pellets or powder are deposited in a flat mold between hot metal plates, which are then pressed together and heated. The plastic melts and solidifies, and is then cooled at room temperature, with pressurized air or in a liquid bath. Figure 2.20 shows the different components of a plastic heat press.

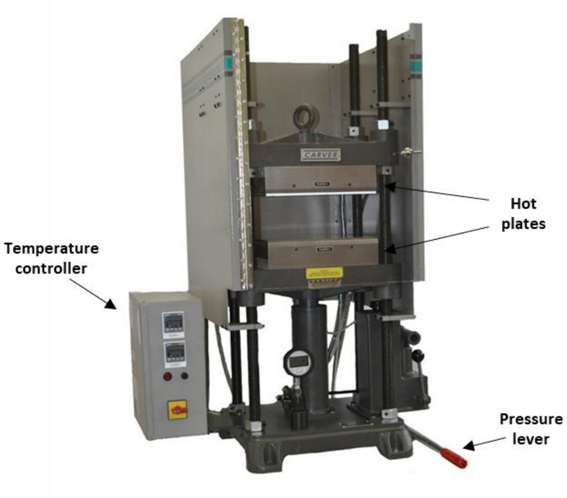


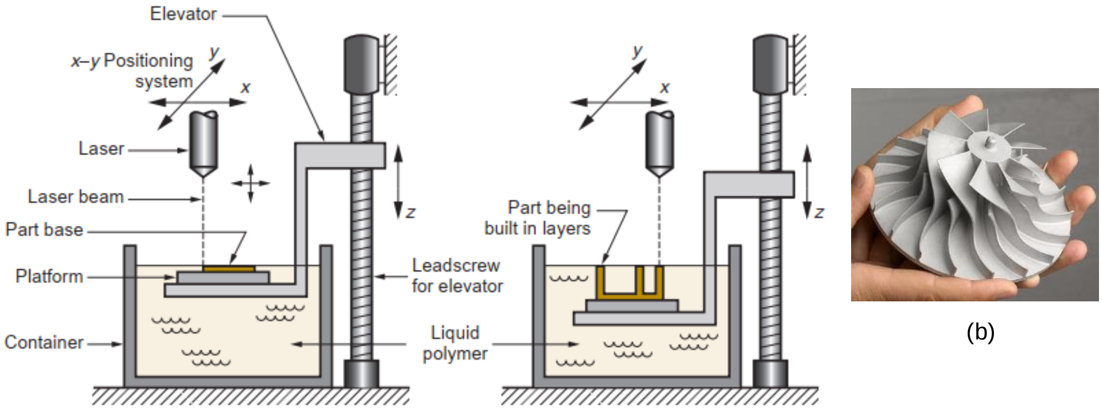
Figure 2.20. Schematics of a plastic heat press (Spectra Services, n.d.)

The pressure is controlled manually with a lever and the temperature is usually controlled electronically. This process is mainly suited for a laboratory setting as it does not offer proper control of parameters for industrial manufacturing.

There are additional plastic manufacturing processes for production of sheets, films and fibers or filaments, but they are typically calendering or a combination of both calendering and blown-film extrusion processes.

Additive manufacturing

This process deposits material layer by layer until a part designed in a computer assisted design (CAD) is formed. Additive manufacturing of plastics can be divided into 3 different processes: fused deposition modeling (FDM), stereolithography (SLA) and selective laser sintering (SLS). Figure 2.21 shows simple schematics for an SLA printer (a) and a part manufactured through additive manufacturing (b).



(b)

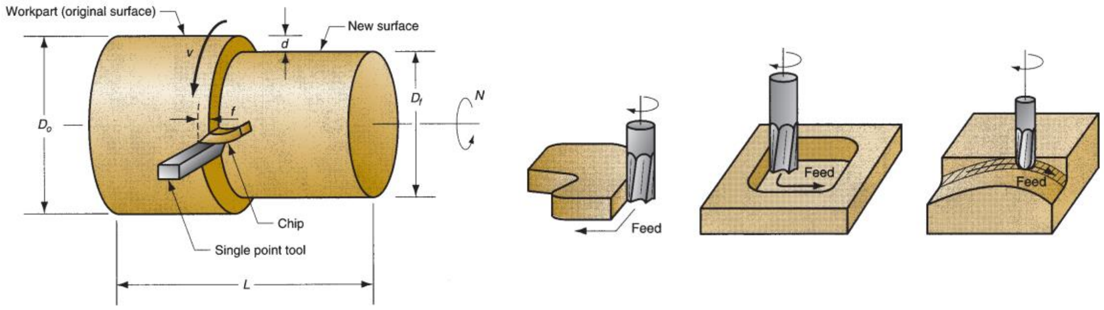
(a)

Figure 2.21. (a) SLA diagram (Groover, 2013) (b) propeller produced by additive manufacturing (3D Systems, 2017)

In FDM, the plastic filament is fed to the machine to be melted, and it is then layered according to the CAD design chosen. As the part is being built, some form of cooling with fans might be done to accelerate the setting process. In SLA a liquid resin is cured, and the shape is formed as it solidifies. SLS is a technique where plastic is fused. being printed, parts often require some form of post processing like cleaning, washing, post-cute or even having support structures removed (often these structures are built in a different material that can be dissolved in a special solution, leaving intact the part's structural material). Additive manufacturing has cost advantages comparing to more traditional manufacturing processes because it requires no tooling and very little lead and set-up time. On the other hand, manufacturing is a lot slower than in other processes, making it generally unsuitable for mass production of parts. Additive manufacturing is generally used with thermoplastics and thermoset resins, depending on the specific process, and can replicate complicated geometries, often with size limitations (the part can only big as big as the scope of the machine). This technology allows designer to quickly produce a part and check for mechanical interreferences with other parts or use the part to support CAD models.

CNC Machining

This process is computer controlled. Similarly, to additive manufacturing, a CAD design is fed to the machine but instead of material being layered, the part is shaped by removing material from a block. For this reason, this is a material removal process. CNC machining can be done with a spinning tool and a fixed part - milling – or a spinning part and a fixed tool – lathing, much like metal parts. Figure 2.22 shows two machining operations that can be performed by CNC machines: turning (a) and three examples of milling (b).



(a)

(b)

Figure 2.22. (a) CNC operations: turning (b) different types of milling (Groover, 2013)

CNC machines require not only a CAD design but also the steps required to produce the part through computer assisted machining (CAM). This process has moderate set up costs and high production costs as it does a long time to produce one part comparing to more traditional processes. CNC machines can produce highly complex parts, as seen in Figure 2.22. The detailed work is ideal for this process, but it does have geometry restrictions and needs allowances for tool access. This can be an issue for internal channels, for example. CNC machined parts usually require some cleaning and trimming after being produced. Due to its nature, CNC machining is more appropriate for harder plastics, including Acrylic (PMMA), PE, PP and Polycarbonate (PC). Softer thermoset plastics can be machined using CNC, but they may require tooling to support the part during machining.

2.1.5. Environmental Impact

There are 3 legal paths generally considered for plastic waste disposal: mechanical recycling, energy recovery and landfill disposal (Gu et al., 2017). Figure 2.23 shows that from all the streams of plastic material, from production to end life, very little is recycled.

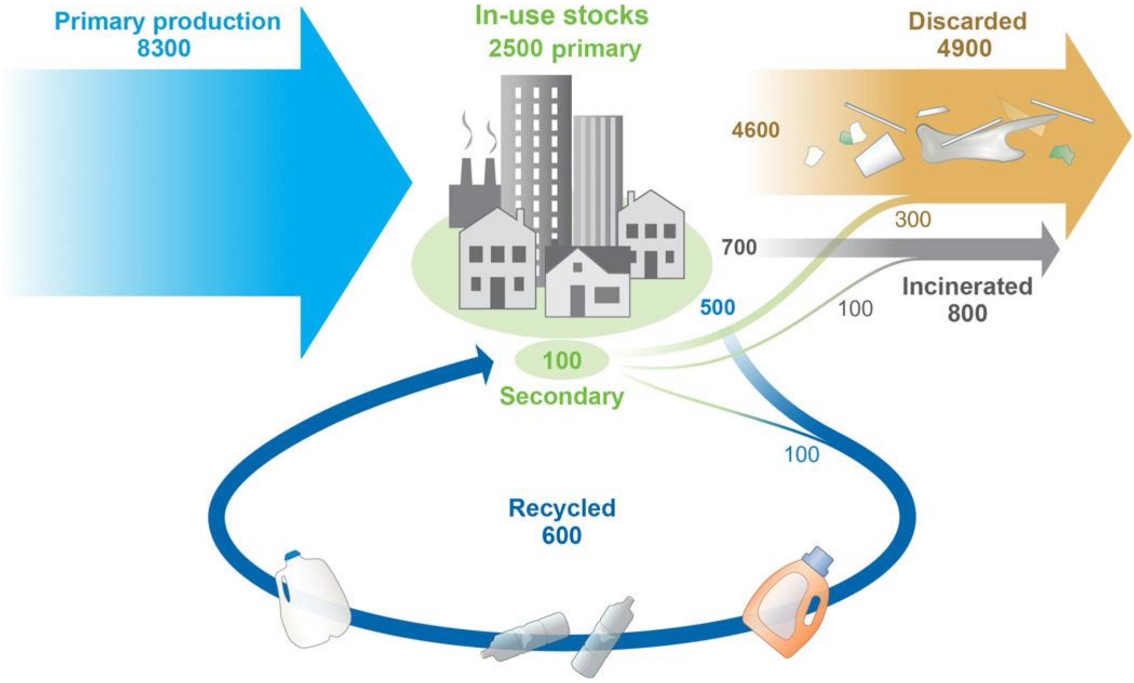


Figure 2.23. Global production, use and fate of polymer resins, synthetic fibers, and additives 1950 - 2015, million metric tons (Geyer et al., 2017)

From all the plastic produced since 1950 to 2015, just over seven percent has been recycled. Notably, as mentioned before, only about 30% of plastic produced is still in-use, supporting the conclusion that most plastic products have relatively short lifespans when comparing to other materials like metals and ceramics (Lebreton et al., 2018). Over half of plastic produced is considered to be “discarded”, which is a general term to explain that it has leaked into land or ocean. It is also worth mentioning that it is hard to accurately estimate the amount of plastic waste leaked to land and especially to the oceans because correct and continuous monitoring has not been consistent since the beginning of plastic production. This issue will be further explored below.

As the CE strategies will be presented next, it is worth considering the opportunity for mechanical recycling of plastic. As seen before, the most common used plastics for packaging, the most used application for global plastic production, are PP, PE and PET. Both thermoplastics are not biodegradable but can be mechanically recycled.

Concerning the environmental impact caused by plastic, production and even “acceptable” disposal methods like energy recovery and landfills, pose risks for human and animal health, as well as for the environment (Thompson et al., 2009). Landfill can have long term environmental effects like contamination of soil and groundwater by additives and by-products of the breakdown of plastics (Hopewell et al., 2009). Incineration presents health and environmental risks because of the possibility of releasing hazardous substances into the atmosphere (Hopewell et al., 2009), as well as CO2 emissions (Eriksen, 2019). An appealing aspect of incineration is that the volume of landfills decreases, but this method does not lessen demand for fossil fuels, as most products incinerated are made from virgin plastic. Additionally, it is worth noting that incineration also prevents SPW of ending up in the ocean.

In a study by (Thompson et al., 2009) data on established knowledge, specific environmental and health concerns, as well as recommendations for industry, research and policy was gathered. Annex A presents this information. Figure 2.24 shows possible pathways for degraded SPW and the potential impacts for wildlife and humans once it enters marine habitats and its food chains.

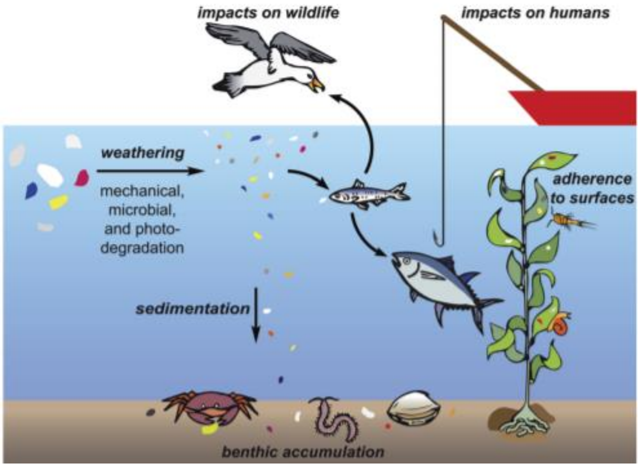


Figure 2.24. Pathways and impact of microplastic in marine environments and humans (Lin, 2016)

Due to weathering effects, SPW breaks down not only into ingestible-size pieces but also into microscopic ones. It is then incorporated by wildlife into food chains, eventually reaching humans.

Studies point to 250 thousand tons of plastic debris floating in the oceans (Eriksen et al., 2014) and an annual leakage of about 8 million tons (Magnier et al., 2019). Another study estimated that SPW makes up to 80% of waste found on land, shorelines, ocean surface and seabeds (Auta et al., 2017; Barnes et al., 2009), making it the most significant source of marine pollution and contamination. The Ellen MacArthur Foundation gauges there will be more plastic than fish in the ocean (by weight) by 2050 (Kaplan, 2016).

Significant contributors to the marine SPW issue are costal human activities. Figure 2.25 shows the relation between coastal population and mismanaged SPW.

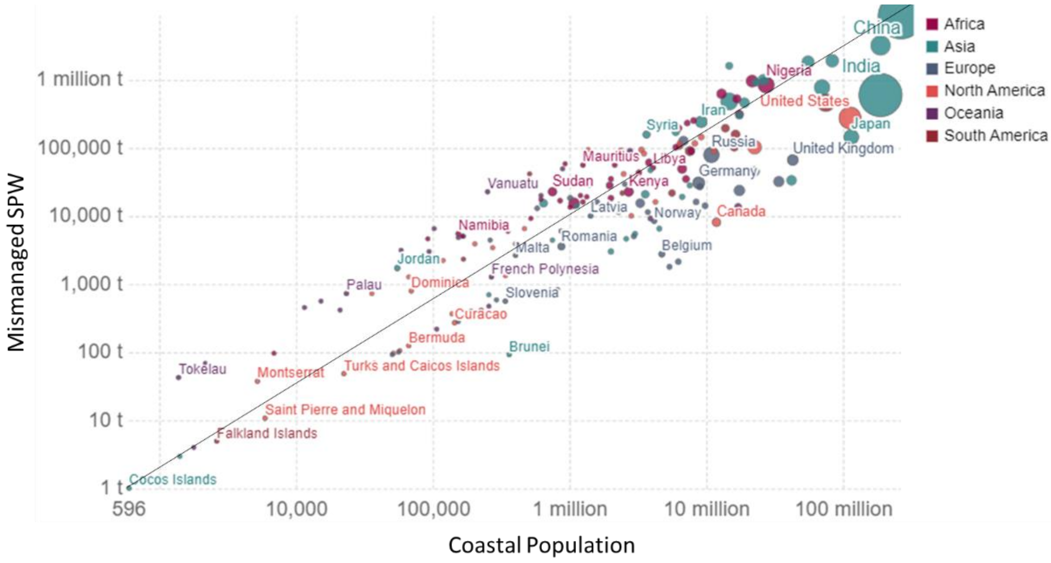


Figure 2.25. Mismanaged SPW measured in tonnes vs. coastal population trend analysis (population within 50 km of the shoreline), adapted from (Ritchie & Roser, 2018)

There is a proportional relation between large coastal populations and mismanaged SPW: the closer populations are to the coast, the higher the amount of mismanaged waste. This issue is not only due to marine commercial activities, like fishing, but also due to leakage through cities' waterways, wastewater outflows and transport through wind or tides.

The location of plastic waste in the ocean is also not exactly known. Again, not only due to insufficient monitoring, but also because plastic degrades and sinks and is consumed by marine organisms making it hard to know its location. Predictions and studies point to buoyant plastic waste converging in subtropical gyres (Borghesi et al., 2016; Eriksen et al., 2014). Gyres are large masses of water circulating in ocean currents, particularly those involved with large wind movements. Figure 2.26 shows the 5 subtropical gyres, with numbers' order and size indicating their size.

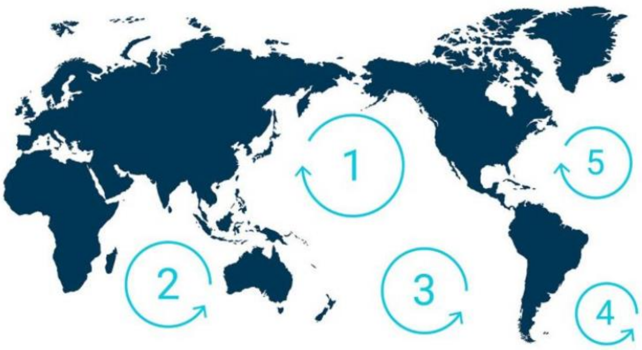


Figure 2.26. Pathways and impact of microplastic in marine environments and humans (Lin, 2016)

Numbers 1 and 3 show the North and South Pacific gyres. Number 2 represents the Indian gyre, and numbers 4 and 5 represent the North and South Atlantic gyres, respectively. The gyres are shown in the figure with their respective rotation direction.

The large concentration of plastic debris is thought to be in the North Pacific gyre (The Ocean Cleanup) midway between Hawaii and California. It is called the Great Pacific Garbage Patch (GPGP). A notable study by Lebreton et al. (2018) aimed to characterize the GPHP. Figure 2.27 shows the sources of plastic in GPGP in 2015.

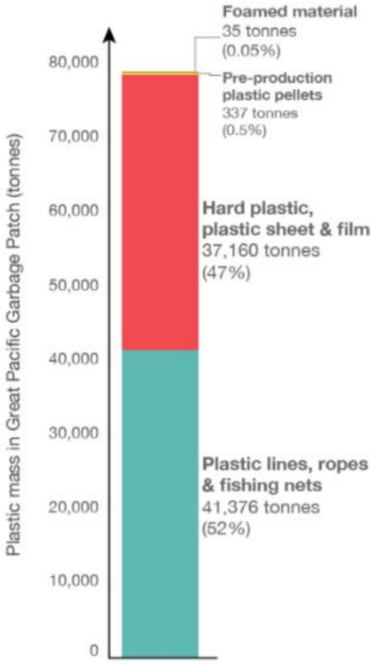


Figure 2.27. Composition of the mass of SPW in the GPGP (Ritchie & Roser, 2018)

This concentration is due to the large coastal population accessing this area and the intensive fishing activities. The area of floating SPW is estimated to be of about 1.6

million km^2 (*The Ocean Cleanup*, n.d.) and its composition is “dominated” by PP and PE, as expected from data of global production and use of these plastics.

Aside from its estimates, the study once again highlights the uncertainty of the data found, saying the actual amount of SPW is likely much higher. This conclusion is due to many reasons. One of them is that large environmental events like tsunamis are believed to have transported a big number of debris directly to the ocean floor (these plastics cannot be assessed through the air, space or water-borne technology used in the study). On the other hand, a general lack of certain products in the samples found, like films and plastic bags, alerted the authors to the likely possibility of sunken SPW (these items have high global plastic production). The study also warns to the possibility of SPW in the GPGP continuing to degrade and sink into the ocean floor or into microscopic pieces.

2.2. Circular Economy

Linear Economy (LE) or industrial economy, is the traditional and most common way of considering economic value of materials and products. This theory considers that to increase value, as much raw material as possible must be extracted, so that the highest number of products can be produced and sold. These products are then disposed of once their lifetime (preferably as short as possible), ends. Disposal is usually of no concern to the elements of the supply chain of the products sold, and the responsibility to ensure it is properly done resides solely on the consumer (Stahel, 2019). Profits for LE current of thinking translate to monetary value, meaning considerations such as natural reserves of material, value of products in use or proper reinsertion of materials transformed are not contemplated (*Circular Economy*, n.d.).

Circular Economy (CE) comes as a response to LE, to preserve value, not only monetary but also of products and materials, as much as possible. It can be quite difficult to define CE as it is a relatively new concept and can be termed an “umbrella concept” (Blomsma & Brennan, 2017) being a somewhat ambiguous term. As a starting point to explaining CE, Table 2.4 presents the main differences between the traditional LE and CE.

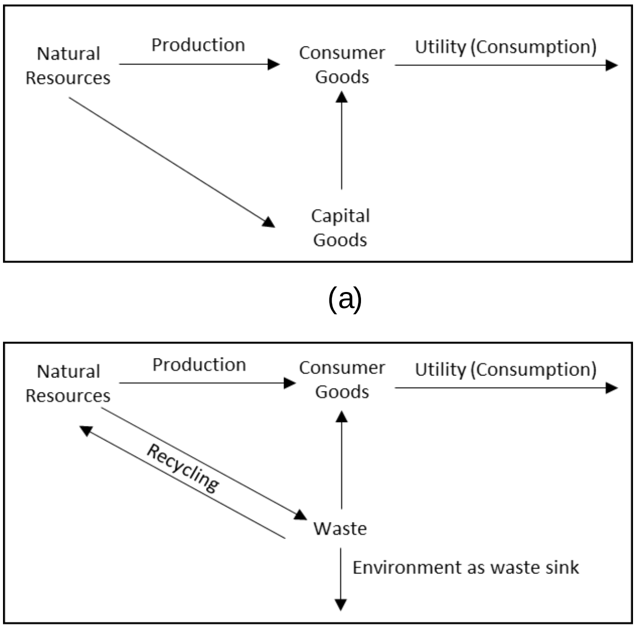
Table 2.4. Main differences between LE and CE

	Circular Economy	**Linear Economy**
Product end of life focus	Designer	Consumer
Business focus	Keep products and materials in use	Produce new products
Environmental dimension awareness	Regenerate natural systems	Use virgin materials
Material flow	Multidirectional	Unidirectional

Both theories are compared regarding their four main differences: product end of life focus, business focus, environmental dimension awareness and material flow.

Design is the first and most important distinction between both approaches because all the steps after that - extraction of materials, production, and sale - assume that the higher the number of products sold the more efficient the business model is. This rationale is opposite to the CE one, where products and resources are meant to keep their value and are ideally used “forever”.

Another important distinction between circular and linear is the material flow perspective, as seen in Figure 2.28.



(a)

(b)

Figure 2.28. (a) Material flow in linear economy (b) circular economy (Andersen, 2007)

While CE aims to optimize circularity and close loops with materials, products and natural resources (multidirectional flow), LE keeps extracting natural resources to produce new products (unidirectional flow).

CE as a loop system was first described by (Stahel & Reday-Mulvey, 1981). Their work was motivated by the need for changes in the way natural resources are introduced and maintained in the economy. They defined CE as a structure and nature of an “economy in loops”.

Because this work aims to apply CE principles to the mitigation of ocean SPW, Annex B details the main drivers identified in literature to enable or hinder CE. These motivations are relevant because to implement CE techniques, it is firstly relevant to identify probable factors that will affect it positively or negatively. The review found that lack of adequate technology, a clear CE framework (as well as a consensual definition of CE itself and its strategies) and standards are dominating barriers to the implementation on CE techniques. While public awareness was identified as a barrier, it also constitutes an enabler – consumer awareness dictates market availability of products containing less plastic or recycled plastic. On the other hand, public health and environmental concerns, as well as capital and policy, were found to make positive contributions in advancing CE strategies.

Although CE is a relatively new concept, the issue of how an overuse of natural resources could strain it to a limit point and jeopardize human evolution had arose much earlier. Most notably, in Malthus’s “An Essay on the Principle of Population”, written in 1798, which would come to influence and shape economy and environmental research for many decades afterwards. Malthusian theory was that a too high increase in population growth would strain natural reserves to the point that food supply would not be enough, and terrible consequences like famine would ensue. This theory was used to support extreme population control theories and practices, but also as the base for CE rationale: natural resources should be preserved in a way that makes for sustainable growth. Other theories and approaches have been developed both before and since, but the term “Circular Economy” was first used by Pearce and Turner (Pearce & Turner, 1989). An overview of notable interpretations of waste and resource management is presented in Figure 2.29.

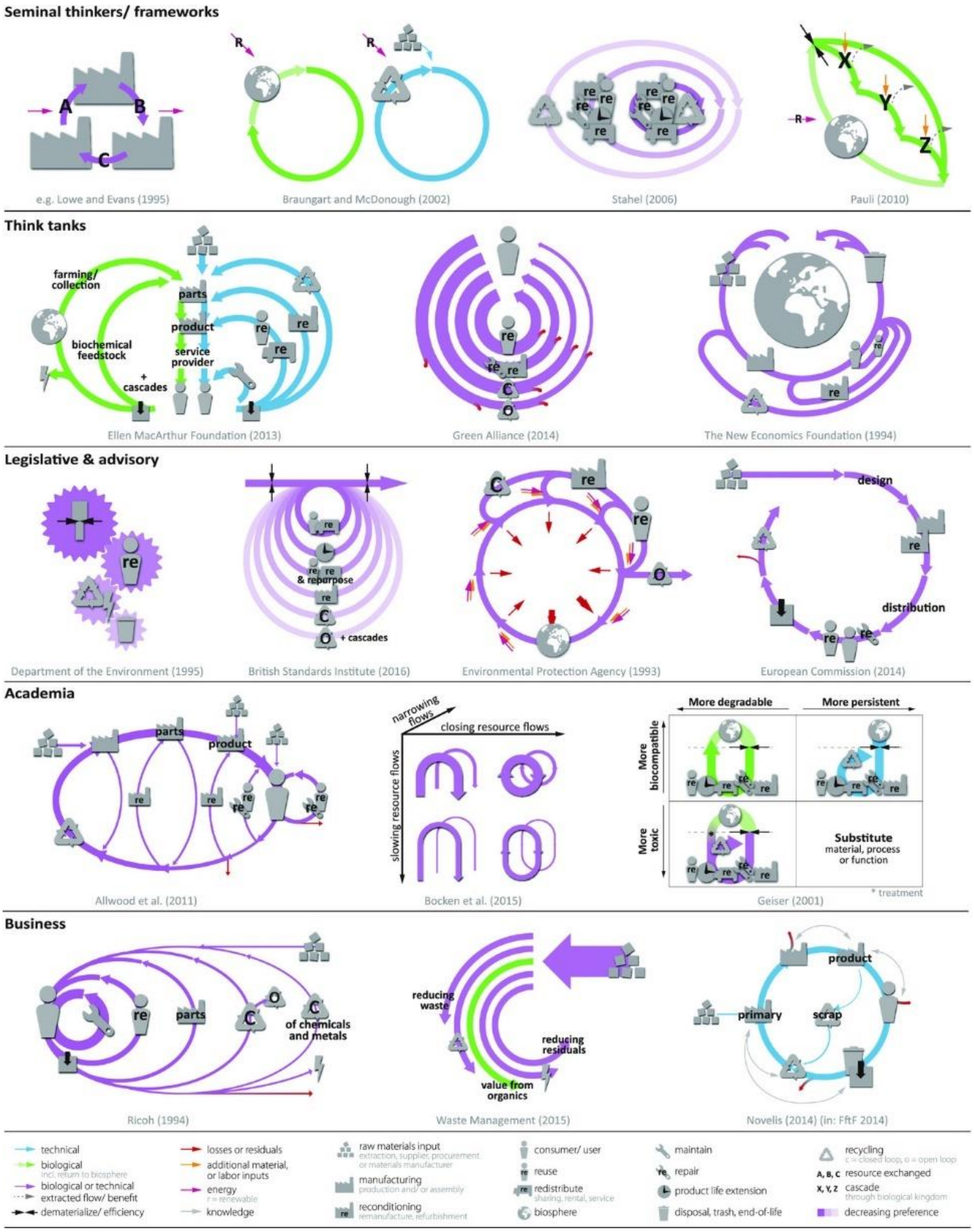


Figure 2.29. Waste and resource management framework (Blomsma & Brennan, 2017)

As explained before, CE is open to interpretation and there are many different focus points to be taken. Depending on the expertise field and focus, different sectors of society tend to think of it in different ways. Common points between most interpretations are that bigger and smaller loops exist and circularity increases as these loops' size decreases. CE definitions aim to model and regulate the way resources, natural, human, or both, flow in systems. It is worth mentioning the "Butterfly Diagram" from the Ellen MacArthur Foundation, which will be further explained in this section. This interpretation is relevant because the Ellen MacArthur Foundation's definition of CE is a consensual one and is the one used in this work.

The Ellen MacArthur Foundation's interpretation of CE aims at reshaping the way finite natural resources are used and renewed, based on three fundamental principles:

- Design out waste and pollution
- Keep products and materials in use
- Regenerate natural systems

To better understand circularity and its strategic options, the Ellen MacArthur Foundation presents the Butterfly diagram, Figure 2.30.

The diagram shows two types of resources, or "nutrients", green and blue. Green nutrients are biological materials and can safely re-enter the natural world. They decompose or are absorbed and transform into usable natural resources. Blue nutrients are technological materials and cannot do the same. Technological nutrients cannot be reabsorbed into biological loops because they have been extensively manipulated through irreversible processing. To prevent the resources used to make technological nutrients from leaking – through solid waste or hazardous contaminations - ways to keep their value must be found.

Plastics are considered technological materials because they do not biodegrade, and because they represent such a large amount of waste, their value or "lifetime" must be extended as much as possible.

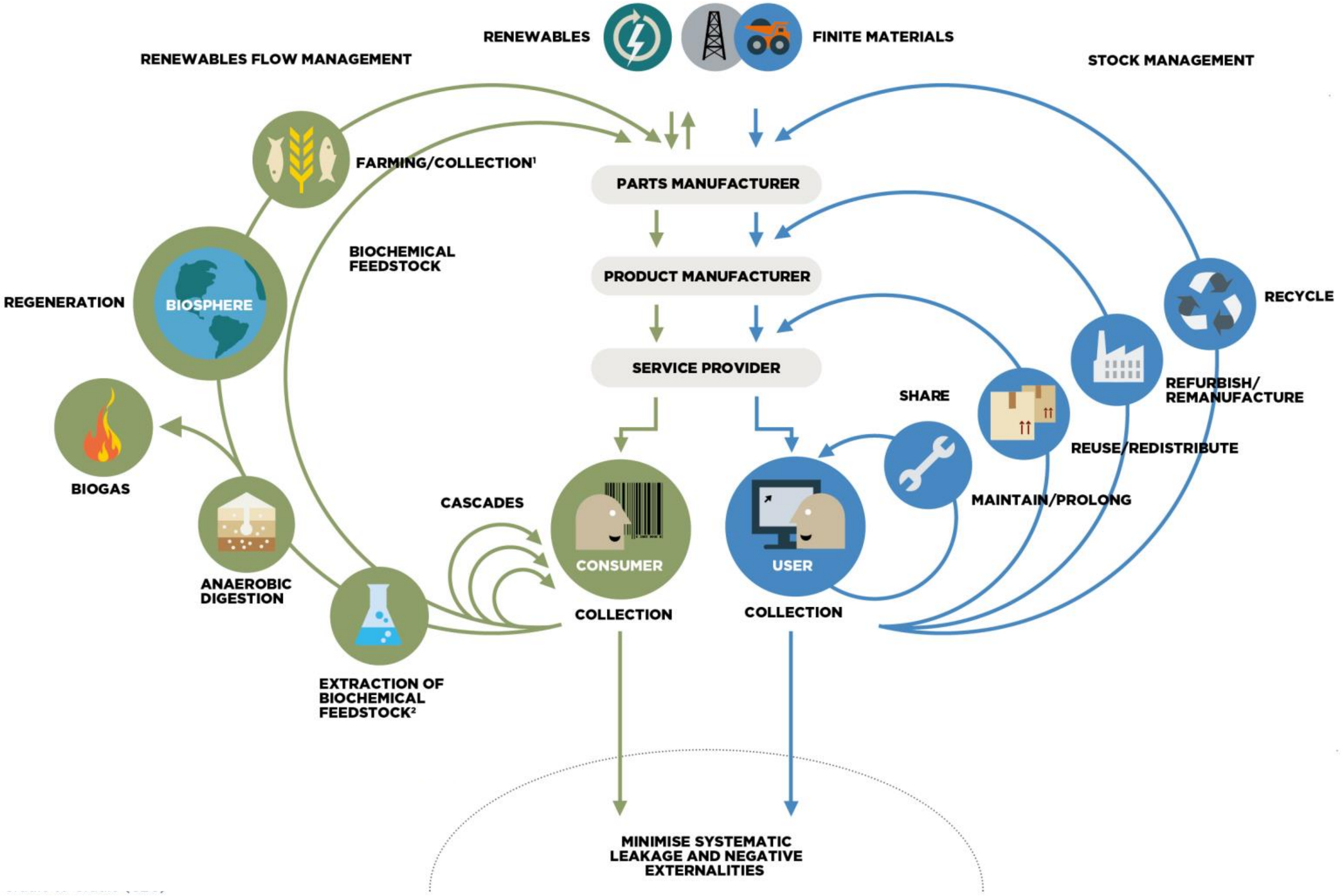
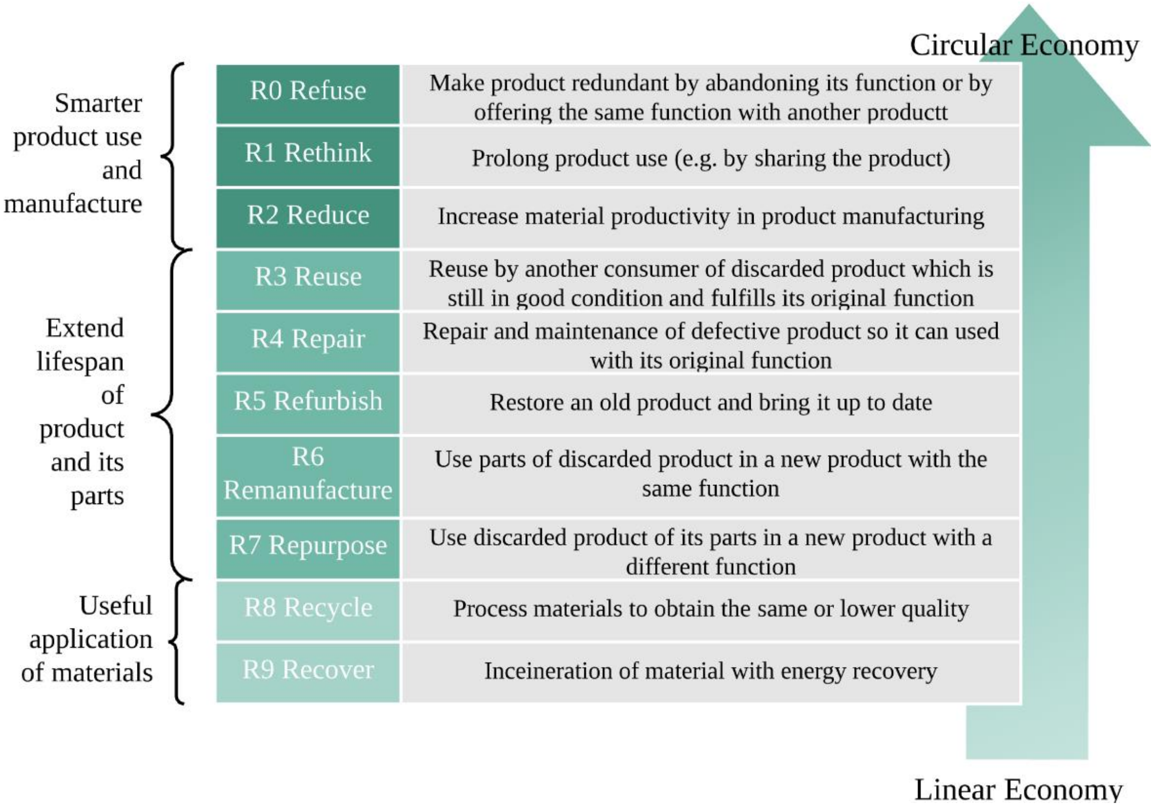


Figure 2.30. The Butterfly Diagram (*Circular Economy System Diagram*, n.d.)

The Ellen MacArthur Foundation has prioritized strategies to achieve circularity in economies in its 9 R’s chart, which can be seen in Figure 2.31.



R0 Refuse	Make product redundant by abandoning its function or by offering the same function with another productt
R1 Rethink	Prolong product use (e.g. by sharing the product)
R2 Reduce	Increase material productivity in product manufacturing
R3 Reuse	Reuse by another consumer of discarded product which is still in good condition and fulfills its original function
R4 Repair	Repair and maintenance of defective product so it can used with its original function
R5 Refurbish	Restore an old product and bring it up to date
R6 Remanufacture	Use parts of discarded product in a new product with the same function
R7 Repurpose	Use discarded product of its parts in a new product with a different function
R8 Recycle	Process materials to obtain the same or lower quality
R9 Recover	Inceineration of material with energy recovery

Figure 2.31. The 9 R's (Potting et al., 2017)

As mentioned above, the best strategies to ensure circularity are the ones that sustain products or materials’ value throughout their lifetime, avoiding consumption of virgin materials, with design at top of the list. However, for the specific case of ocean plastic waste, made up mostly of packaging, mechanical recycling can pose as readily available technology. It is worth noting that plastics’ value is very difficult to recover due to its complicated manufacturing nature and applications.

Under a CE perspective, recycling ocean plastic waste would be both a lucrative opportunity as well as a chance to mitigate a considerable environmental issue, effectively creating a significant stream of valorization of plastic waste. Recycling plastic waste should be more economic than processing virgin materials and while this is globally true, local issues might obstruct financial gains. For example, it can be too expensive to transport waste to a recycling plant, or policymakers might favor the production of new products. Stahel argues that high-volume, low-value recycling is widely used to either minimize costs or because proper recycling technology is unavailable. Nevertheless, the extraction of natural resources through mining and further processing inevitably costs more as a whole, in the long run. These costs are not only financial – be it through energy and water consumption as well as waste generated disposal – but also environmental. It has been documented that mechanical recycling has less Greenhouse Gases (GHG) emissions than production of virgin plastic (Ragaert et al., 2020). In conclusion, financial opportunities for recycling should not be ignored in favor of faster gross domestic product (GDP) growth.

In summary, to close technical loops for plastic, mechanical recycling is a viable solution and for this reason, it is further explored in the next section.

2.3. Recycling

There are three main types of recycling: mechanical, chemical and energy recovery. A brief description and comparison of nomenclature used in various sources (Baytekin et al., 2013; Hopewell et al., 2009; Maris et al., 2018; Ragaert et al., 2017; Singh et al., 2017) for the different techniques is shown in Table 2.5. CE categorization refers to the application of the recovered material: if it can be maintained long in use then it is closed-loop, but if it can no longer serve the same purpose (even if the new one does not necessarily have “lower value”), then it is classified as open-loop recycling, (Ragaert et al., 2017). It is worth noting that closed-loop recycling does not necessarily imply that virgin material is not added. Often recycled plastic is mixed with virgin plastic or in wood composites, to improve its mechanical and physical properties.

The table shows equivalent terms to refer to the same process of recycling according to the ASTM and ISO standards. The processes are ordered from the one which can recover the most value out of the material – mechanical recycling – to the one which can recover less – energy recovery. Although the different standards analyzed in Table 2.5. consider incineration as a “recycling method”, this is usually not the case under a CE perspective, (as seen on Figure 2.30).

Table 2.5. Description and a general comparison of recycling techniques adapted from Hopewell et al., 2009

Recycling technique		Equivalent terms	CE categorization	Description	Advantages	Challenges
ASTM D5033	Equivalent ISO 15270					
Mechanical recycling	Primary recycling	Closed-loop recycling Re-extrusion Re-use	Closed-loop or open-loop	Mechanical reprocessing into a product with equivalent properties	High value recycling Cost effective, efficient and Well-established	Efficiency determined by input plastic waste Deterioration of properties of the material Pre-treatment requirements Not feasible for contaminated or mixed waste (immiscibility of polymer blends)
	Secondary recycling	Downgrading	Open-loop	Mechanical reprocessing into a product with lower properties		
Chemical Recycling	Tertiary recycling	Feedstock recycling Biological recycling Organic recycling	Closed-loop or open-loop	De-polymerization to chemical constituents or composting of polymers	Suited for heterogeneous and/or contaminated plastic waste, difficult or uneconomical to separate Reduces the amount of chemicals required to produce fuels and virgin plastics Operational for PET Simple technology Generates pure value-added products	Mainly limited to condensation polymers Requires high volumes to be cost-effective
Energy recovery	Quaternary recycling	Valorization	Open-loop	Incineration	Suited for all plastic types Generates energy from polymers (equivalent to burning oil)	Potential release of hazardous substances into the atmosphere No net positive energy balance

While mechanical recycling is the process that can retain the most value out of plastics, it is highly dependent on the input plastics (sorting and separating and pre-processing). Chemical recycling can be process mixed plastics, but it is a highly energy intensive process and requires large volumes of material to be a profitable operation. Comparing the three processes, energy recovery is the least preferable one because not only because the gain it generates is not significant – energy obtained is equivalent to that of burning oil and so more energy is spent on the processing than gained from it – but because it can have potential hazardous emissions into the atmosphere. The reason why this method is so use despite its inefficiencies is because it decreases volume of waste in landfills. For each type of recycling there are several different processes to accomplish recycling.

In quaternary recycling (energy recovery), materials cannot be recuperated, only the energy from combustion can be used. With tertiary recycling (chemical recycling), the process needs to be tailored to the polymers composing the plastics to be processed. This can make chemical recycling a process too difficult to implement for bulk recycling of ocean plastic waste. On the other hand, different processes have different circularity measures, and this too is a factor in choosing which type of recycling to choose. Figure 2.32 shows the relative loop sizes for different recovery strategies.

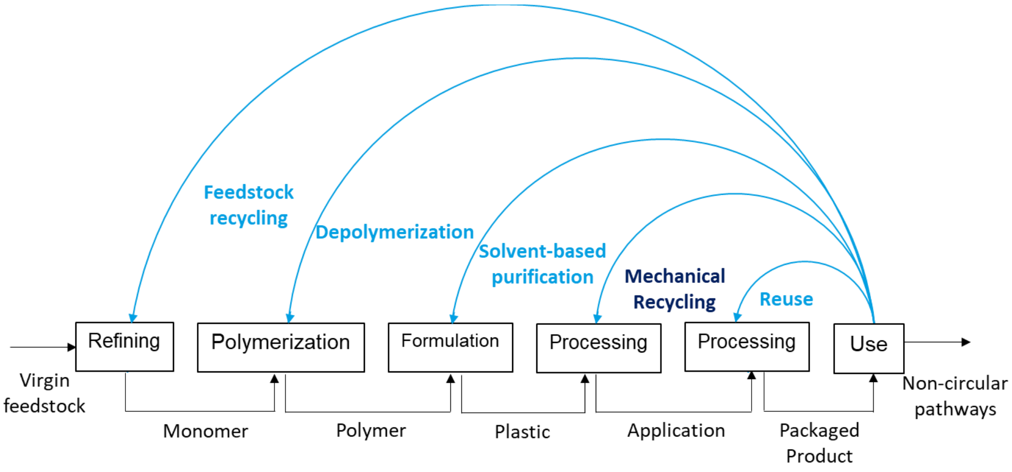


Figure 2.32. Different recovery techniques and their “circularity”, adapted from Crippa et al., 2019

Notably, mechanical recycling is the next best strategy after reusing plastic products. This is because it is the recovery strategy that retains the highest value for plastics (Al-Salem et al., 2009). It is also the most technologically developed and economic of all recycling strategies other than energy recovery (Soto et al., 2018). Bigger loops such as solvent-based purification, depolymerization and feedstock recycling are all chemical recycling strategies. In this model energy recovery is considered a non-circular pathway because the materials loose too much value through this method.

For these reasons, the next section focuses exclusively on mechanical recycling and its surrounding issues.

2.3.1. Mechanical recycling

Mechanical recycling is the most widespread, technologically advanced, economic recycling strategy other than energy recovery. Additionally, mechanical recycling of plastics is the preferred strategy by the EU to implement a more circular economy of these materials (Lazarevic et al., 2010).

Mechanical recycling methods vary but in general the different steps have the same goals (Al-Salem et al., 2009; Ragaert et al., 2017):

- Separating and sorting – separating items according to different characteristics like size, shape, density, and color.
- Downsizing – reducing plastic waste size through cutting or shredding.
- Contaminant separation – removing paper, dust, and impurities in a cyclone.
- Floating – separating different plastics in a tank according to their density.
- Milling – milling of single-polymer plastics. This is often the first step for many recycling plants, decreasing quality the recovered plastic.
- Washing and drying – first stage water washing which is then followed by further washing, either with water or by use of chemicals like caustic soda and surfactants.
- Agglutination – either storing of the resulting material or further processing with pigments and/or additives.
- Manufacturing (Extrusion/Injection) – extruding the plastic to strands which can then be pelletized and reinjected to manufacture single-polymer plastic.
- Quenching – water-cooling of plastic to be granulated.

It is worth noting that how these steps happen – in which order, multiple times or at all – depends on composition and origin of the plastic waste. A simplified scheme of a mechanical recycling process is shown on Figure 2.33.

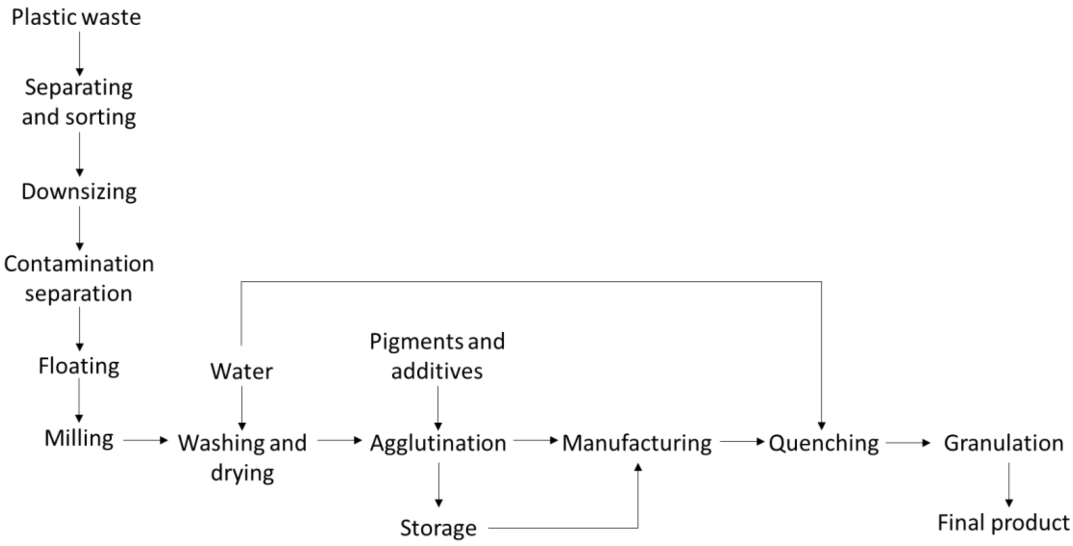


Figure 2.33. Simplified stages mechanical recycling steps (adapted from (Ragaert et al., 2017) and (Aznar et al., 2006))

Mechanical recycling is very much dependent on contamination and degradation of properties throughout the supply chain, as well as a careful optimization of processing parameters during manufacturing. Figure 2.34 shows the potential losses of material and properties since SPW is "generated" until it can be recovered.

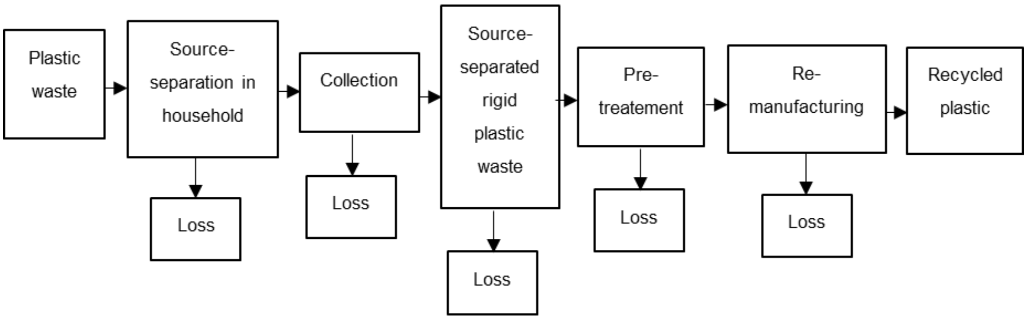


Figure 2.34. Simplified stages mechanical recycling steps (adapted from (Aznar et al., 2006))

The main stages identified in literature as to having influence on performance of recovered polymers are collection, sorting or separation and manufacturing. Because the focus of this work is manufacturing, the next section presents a more detailed analysis of this issue.

Overall results of mechanically recycled plastics found in literature have been satisfactory meaning mechanical properties were equivalent to the ones found in virgin plastics. Variability was found in the results, but all case studies analyzed found that recovered polymers had potential for being applied in the industry, even when mechanical properties of recycled plastics were lower than those of their virgin counterparts (Dahlbo et al., 2018). In general, efficiencies of mechanical recycling are obtained when compared to the polymers' virgin equivalents. It is worth noting that a more detailed analysis for substitution potential of virgin by recycled polymers should be done on a case-by-case basis as design requirements are product dependent and what might not be suitable for one application can be to other(s).

2.4. Variability of recycled plastic's mechanical properties

There is relevant variability in recycled plastics' mechanical properties and this unpredictability is one of the reasons why its use is not more widespread. There are two main reasons for this issue: not only are there significant differences in the quality of recovered plastics, but also several processing issues can arise throughout the supply chain.

Contamination along the supply chain of recovered plastics makes it very difficult to sort "impure" plastic products (Aznar et al., 2006; Ragaert et al., 2017). There is also great degree of variability in both post-industrial and post-consumer waste (PCW). The number of different origins and possible pathways make for a highly variable degree of properties in recovered plastic and impurities found. This issue is especially critical in PCW because it is not as regulated and monitored as the industrial one. Another notable aspect is the economic one. Industrial waste is usually of high quality and is immediately shredded and reused within the same location or sold at a price close to the one of virgin plastic to other businesses to save costs.

Impurities are a factor that greatly contributes to the variability of properties of recovered plastic. Tainting of waste can include other polymers, non-polymeric impurities, additives to improve properties, glues and metals used for coatings or paints. (Pivnenko et al., 2015) summarized sources of contamination and potential for improvement, as seen in Table 2.6.

Table 2.6. - Sources of contamination (C) and potential for improvement (I) (Pivnenko et al., 2015)

Sources of contamination	Supply chain stage							
	Extraction	Production	Manufacture	Use	Segregation	Collection	Sorting	Re-processing
Polymer cross-contamination			C + I		C + I	C	C + I	I
Additives		C + I	C + I					
Non-polymeric impurities			C + I	C	C + I		I	
Degradation			C	C				C + I

Noting that the supply chain for plastics is complex and involves several parties, Table 2.6. highlights the need to monitor all stages of plastics' lifetime.

- Polymer cross-contamination (Eriksen 2019) – because different polymers have different chemical structures, they cannot necessarily be mixed in mechanical recycling. Manufacturing of plastics is often done by injection and in addition to chemical structures, melting temperatures vary making processing of mixtures challenging. And while cross-contamination can take place due to miss sorting, some plastic products are made of more than one type of polymer, which can possibly not be mechanically separable. These cases likely lead to contamination. Table 9 presents an estimate by Eriksen (2019) of European household waste plastic products' composition. It can be concluded that packaging, a significant amount of plastic waste is often made of more than one type of polymer, making polymer cross-contamination a relevant concern.

- Non-polymeric contamination – presence of non-polymeric materials may include products wrongly sorted as plastic (in the household), organic residues from use, metals from coatings or paints and additives (Eriksen, 2019). These materials may include heavy metals and degrade into hazardous substances, like chromium (Pivnenko et al., 2015). While recycled industrial waste has levels comparable to virgin plastic, household waste presents much higher levels, probably due to use and collection contamination. Colored plastic is a simple example of a reason as to why mechanical recycling efficiency can easily decrease: if the products are not treated first to remove inorganic pigments containing heavy metals (Järup, 2003) – right away decreasing the amount of potentially recovered material – these substances can migrate and not only be potentially hazardous but also decrease both mechanical properties and value of the recovered polymer. This is because recovered polymers will take on a "dirty" color, making it harder to work it into sellable plastic commodities (Faraca & Astrup, 2019).

- Degradation of properties – degradation of plastics is characterized by chemical or physical change due to environmental factors (Shah et al., 2008). This means a deterioration of functionality caused by chemical, physical or biological reactions, which in turn causes chain scission and is followed by chemical transformations (formation of structural inhomogeneities) (Shah et al., 2008; Vilaplana & Karlsson, 2008). Degradation can include physical (crystallinity, melting behavior, morphology, thermal history and viscoelastic behavior (Vilaplana and Karlsson 2008)), chemical (breakage or formation of new functional groups) (Pospíšil & Nešpůrek, 1997), optical or electrical properties by means of crazing, cracking, erosion, discoloration, phase separation or delamination. External conditions can include moisture, heat, oxidation, light, ionic radiation, hydrolysis, and mechanical shear (Faraca & Astrup, 2019; Ragaert et al., 2017). Degradation of properties takes place not only during lifetime but also throughout reprocessing. During recovery, thermal and mechanical degradation can also occur (Dintcheva et al., 1997).

As mentioned before, the success of mechanical recycling is very much dependent on contamination and degradation of properties throughout the supply chain, as well as a careful optimization of processing parameters during manufacturing. Annex C provides a review of the literature of different valorization techniques (Gu et al., 2017) used in mechanical recycling. These methods are often used together and can complement each other in achieving high performance recovered polymers.

2.5. Mechanical characterization methods

There are no specific standards to perform mechanical characterization of recovered plastics, they are the same as for virgin ones (ASTM International, 2014). The different tests/standards to be used mostly depend on the application. A standard test for mechanical characterization of plastics is tensile testing.

Other mechanical testing found in case studies of different recycled plastics also included flexural, impact strength such as Charpy impact tests, and fatigue analysis, for example creep stress, deformation at creep point and at resilience point, and nominal deformation at breaking point (an overview is provided in Annex D).

Additional testing required specifically for recovered plastics is usually for either or both of two reasons: unknown characteristics of the recovered material or/and comparison with virgin equivalents. This testing can include MFI and impurities' analysis as well as investigation of potential degradation of properties through thermal degradation and moisture content.

In fact, recycled materials are often used in industrial applications but its percentages in mixtures with virgin materials are reached empirically: some testing is done for a specific part or component and a visual inspection dictates a "safe" amount of recovered material to use.

The lack of standards for mechanical testing, not only of recycled plastic, but also of its mixtures with virgin ones, diminishes widespread industrial use of recycled plastics. For this reason, there is a need for such a method. In the next section, mechanical characterization of mixtures of different percentages of recycled and virgin plastics is presented in the next section.

2.6. Mixtures of recycled and virgin plastics

To maximize the use of recovered plastics, it can be helpful to test mixtures of the recycled material with other materials, usually virgin, that can improve its mechanical properties. The added materials may or may not be other plastics. This is done because although a high-content mixture of recycled plastic might not be suitable for all applications, it can be to some. Assigning different applications to different recovered-plastics content mixtures is one way to assure widespread use of this material and a decrease of virgin ones. Testing different mixtures is also a way to find if mechanical behavior is the same, even if properties decrease with added content of recovered material. In general, efficiencies of mechanical recycling are obtained when compared to the polymers' virgin equivalents. Annex D presents a review of case studies of recycled polymers, the type of testing performed, and the summarized conclusions obtained. Generally, results found that pure recovered materials and their mixtures had equivalent properties to those of virgin materials.

Soto et al. (2018) studied mechanical properties of different PE products, recycled and in a mixture of 75% of recycled and 25% of virgin material. Although the reasoning for this specific ratio was not presented, it can be speculated that it is approximately the one used empirically industrial applications to cut costs and assure proper quality of plastic products. Figure 2.35 shows some mechanical properties obtained for the mixtures and recycled material.

Tensile properties of recycled film.

Direction	Sample	Creep stress σ_y (MPa)	Deformation at the creep pointε_y (%)	Resilience σ_M (MPa)	Deformation at the resilience point ε_M (%)	Nominal deformation at the breaking point ε_B (%)
Parallel	Recycled PE	–	–	16.1 ± 1.7	362 ± 75	375 ± 69
Perpendicular		8.4 ± 0.3	8.7 ± 0.5	12.4 ± 1.4	617 ± 61	622 ± 60
Parallel	75% recycled PE/25% virgin PE	–	–	17.7 ± 0.3	110 ± 15	138 ± 21
Perpendicular		8.3 ± 0.2	8.2 ± 0.4	12.5 ± 1.3	590 ± 62	594 ± 60

ε = strain and σ = stress.

(a)

Tear resistance of recycled film.

Direction	Sample	Force (N)	Tear strength (N/mm)
Parallel	Recycled PE	2.01 ± 0.16	34.9 ± 4.5
Perpendicular		*8.86 ± 0.41*	*147.7 ± 6.9*
Parallel	75% recycled PE/25% virgin PE	0.55 ± 0.05	9.8 ± 1.0
Perpendicular		*8.70 ± 0.30*	*150.1 ± 4.9*

Data in italics are not reliable.

(b)

Figure 2.35. Tensile properties (a) and tear resistance properties (b) of both a recycled and a 75%-25% mixture of recycled and virgin PE: t (Soto et al., 2018)

Properties found for both the recycled material and the mixture were found to be equivalent to those of virgin PE. Additionally, a slight negative effect was found in the mixture comparing to the 100% recycled material.

In another study, (Sommerhuber et al., 2015) studied the possibility of turning milk bottles from PCW, made from different grades of PE, into wood-plastic composites. Figure 2.36 shows a comparison for Modulus of Elasticity of the different mixtures used.

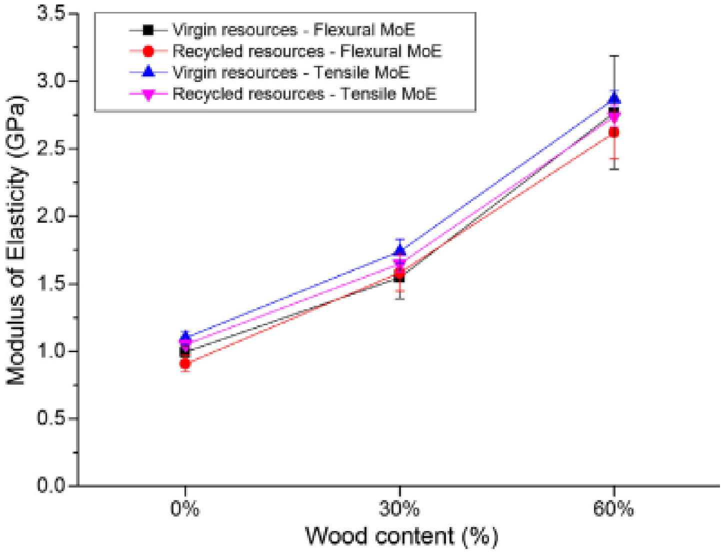
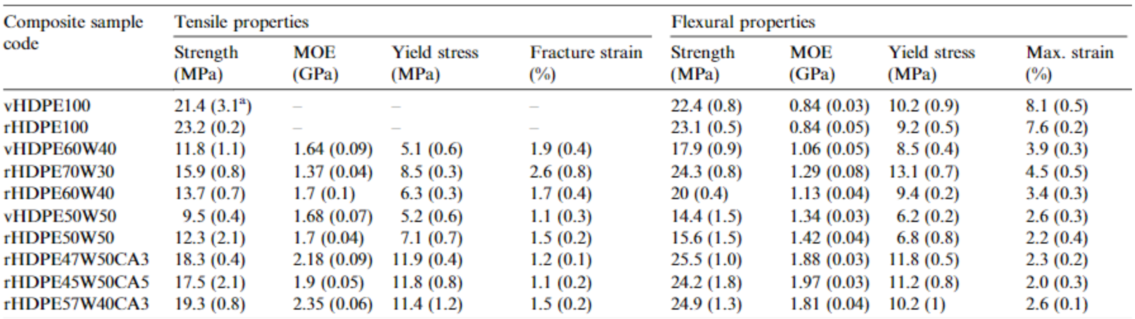


Figure 2.36. Flexural and tensile modulus of elasticity of PE and HDPE and wood mixtures (Sommerhuber et al., 2015)

The so-called "recycled resources" included high-density polyethylene (HDPE), maleic anhydride polyethylene (MAPE), and wood. The "virgin resources" were the equivalent HDPE ad MAPE (chosen according to their recycled counterparts' MFI) and virgin wood.

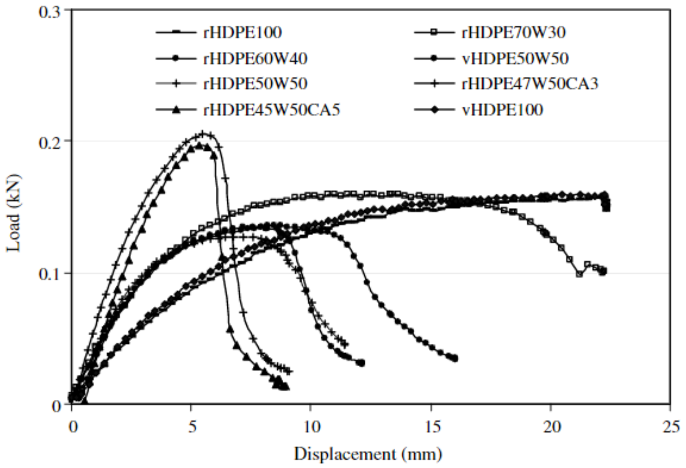
After performing tensile, flexural and Charpy impact tests, it was concluded that properties of recycled plastics were comparable to those of the virgin ones. Additionally, mixtures with a low content of wood (30%) showed no significant differences between recycled and virgin materials' properties.

In another study with sawdust and plastic composites (in which mixtures of only recycled and virgin plastics were also analyzed), (Adhikary et al., 2008) tested samples for physical properties such as density and moisture absorption, and mechanical ones (strength, Young Modulus, yield stress, fracture, and maximum strain). Figure 2.37 shows the properties for the different mixtures and the plot of load – displacement.



Composite sample code	Tensile properties				Flexural properties			
	Strength (MPa)	MOE (GPa)	Yield stress (MPa)	Fracture strain (%)	Strength (MPa)	MOE (GPa)	Yield stress (MPa)	Max. strain (%)
vHDPE100	21.4 (3.1[a])	–	–	–	22.4 (0.8)	0.84 (0.03)	10.2 (0.9)	8.1 (0.5)
rHDPE100	23.2 (0.2)	–	–	–	23.1 (0.5)	0.84 (0.05)	9.2 (0.5)	7.6 (0.2)
vHDPE60W40	11.8 (1.1)	1.64 (0.09)	5.1 (0.6)	1.9 (0.4)	17.9 (0.9)	1.06 (0.05)	8.5 (0.4)	3.9 (0.3)
rHDPE70W30	15.9 (0.8)	1.37 (0.04)	8.5 (0.3)	2.6 (0.8)	24.3 (0.8)	1.29 (0.08)	13.1 (0.7)	4.5 (0.5)
rHDPE60W40	13.7 (0.7)	1.7 (0.1)	6.3 (0.3)	1.7 (0.4)	20 (0.4)	1.13 (0.04)	9.4 (0.2)	3.4 (0.3)
vHDPE50W50	9.5 (0.4)	1.68 (0.07)	5.2 (0.6)	1.1 (0.3)	14.4 (1.5)	1.34 (0.03)	6.2 (0.2)	2.6 (0.3)
rHDPE50W50	12.3 (2.1)	1.7 (0.04)	7.1 (0.7)	1.5 (0.2)	15.6 (1.5)	1.42 (0.04)	6.8 (0.8)	2.2 (0.4)
rHDPE47W50CA3	18.3 (0.4)	2.18 (0.09)	11.9 (0.4)	1.2 (0.1)	25.5 (1.0)	1.88 (0.03)	11.8 (0.5)	2.3 (0.2)
rHDPE45W50CA5	17.5 (2.1)	1.9 (0.05)	11.8 (0.8)	1.1 (0.2)	24.2 (1.8)	1.97 (0.03)	11.2 (0.8)	2.0 (0.3)
rHDPE57W40CA3	19.3 (0.8)	2.35 (0.06)	11.4 (1.2)	1.5 (0.2)	24.9 (1.3)	1.81 (0.04)	10.2 (1)	2.6 (0.1)

(a)



(b)

Figure 2.37. Tensile and flexural properties (a) and load-displacement comparison (b) of different mixtures of recycled and virgin HDPE and wood composites (Adhikary et al., 2008)

The mechanical properties of the mixtures with recycled HDPE were found to be similar or better than the ones for the mixtures using virgin HDPE, namely tensile strength (compared for samples of 100% recycled and 100% virgin HDPE). Additionally, it can be concluded from the load-displacement plots that recycled and virgin composites have the same mechanical behavior.

3. Materials and methods

To assess the methodology described in section 3.3., different mixtures of recycled and virgin plastics test specimens were developed and tested. This chapter firstly describes the mechanical testing performed, namely tensile testing. The properties of all materials used as well as the geometry and size of the pellets used during manufacturing are presented. Afterwards, the test specimens' manufacturing and issues found during this stage, as well as additional testing to try to produce better specimens, are described. Finally, the experimental plan detailing different manufacturing methods, as well as the groups of test specimens compared, is shown.

3.1. Mechanical characterization of materials

To find the mechanical properties of the recycled materials, and compare them to their virgin equivalent, mechanical testing needs to be performed. There are several mechanical tests with different scopes and objectives, and they should be chosen keeping in mind the overall conditions the part will be subjected to. Regardless of the specific application, some mechanical tests are very comprehensive and should likely be performed. This is the case with tensile testing, which is considered the most commonly used mechanical test, which allows to determine essential mechanical properties of materials, and also perform quality control once these are transformed into parts (Martins & Rodrigues, 2010).

Keeping in mind a general approach to the use of recovered plastics, and the general character of uniaxial tensile testing, this was the test performed and its main aspects are further explored in the next section.

3.1.1. Tensile test

The uniaxial tensile test is characterized by applying a continuously increasing tensile load to a test specimen until it fractures. Test specimens used in this test can have either a rectangular or circular cross section. The standard followed for the manufacturing of the tensile tests was the ASTM D 638 – 02 ('Standard Test Method for Tensile Properties of Plastics', 2002), and type IV specimen geometry was selected, as shown in Figure 3.1. This geometry was chosen as it is the preferred on to compare different polymers.

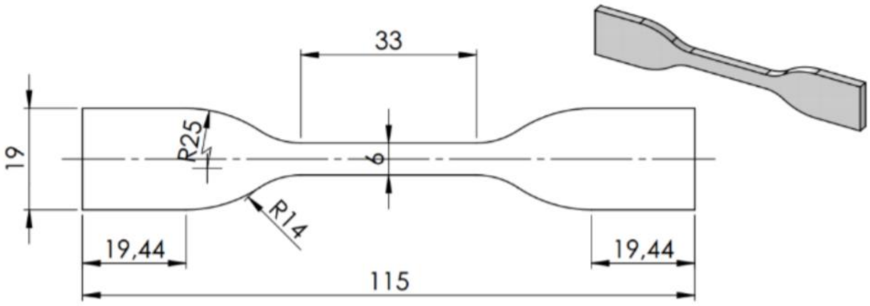
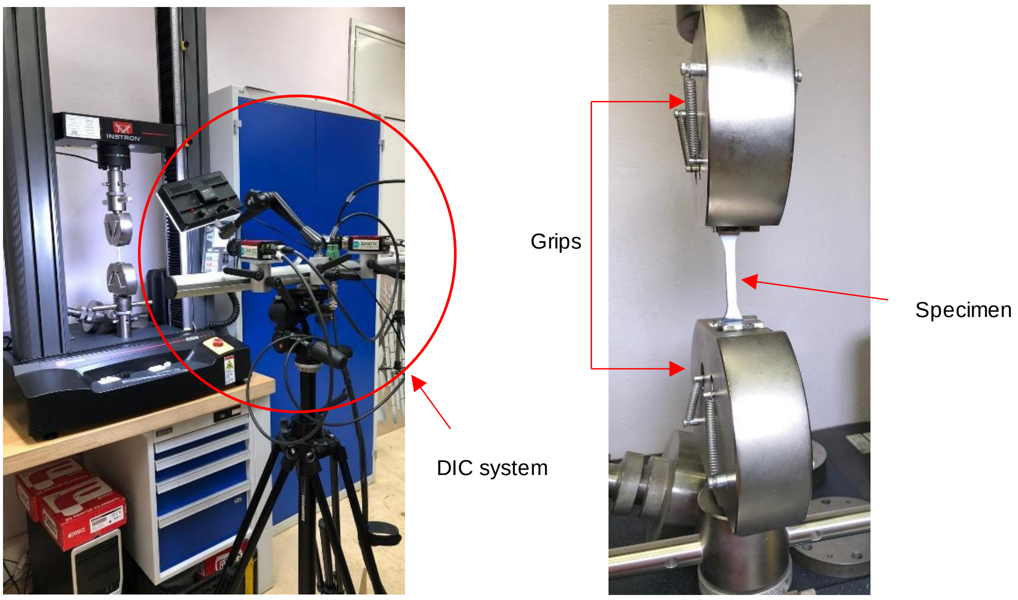


Figure 3.1. Dimensions of the type IV test specimens according to the ASTM D 638 - 02 standard

The tensile tests were performed in the Mechanical Testing Laboratory of the Scientific Area of Manufacturing and Industrial Management. The INSTRON 5966 machine was used, and a digital image correlation (DIC) system was used to measure the deformation of the test specimens. Figure 3.2 shows the set-up used and a specimen being tested.



(a) (b)

Figure 3.2. Set up used for the tensile tests with the DIC system (a) and detail of a specimen being tested (b)

The parameters considered for the tensile tests are the following:

- Testing velocity: $5\ mm/min$, according to the ASTM 638 D – 02 standard
- Data acquisition rate: $0.1\ /s$
- Room temperature: $\sim 20\ °C$

The output from the Instron machine was the following:

- Time $t\ [s]$

- Load F $[N]$
- Displacement $[mm]$

The DIC system used was the Q-400 3D by Dantec Dynamics model, and the gauge length area of the test specimens was painted in white and a black speckle pattern to allow the software to compute the deformation, as shown in Figure 3.3.

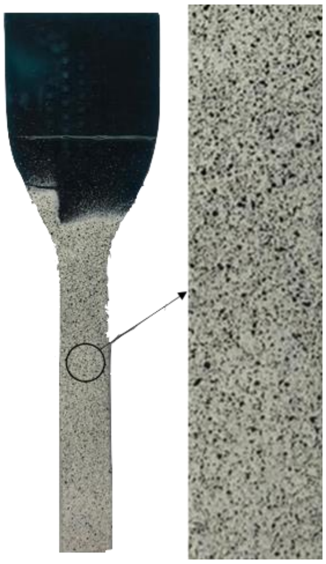


Figure 3.3. Painted test specimen (a) and detail of the painted pattern (b)

For the mechanical characterization of the test specimens two points were selected distanced of the initial gauge length of 25 mm, see Figure 3.4, and the DIC system was used to find the displacement of the two points along the deformation.

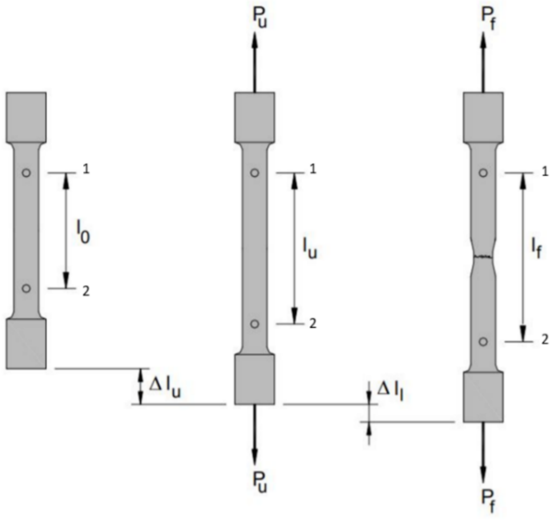


Figure 3.4. Displacement measured between 2 points in a specimen (adapted from (Martins & Rodrigues, 2010))

The deformation between the two points in the test specimens, allowed to obtain the variation of the gauge length, Δl.

To compute the data for the mechanical characterization additional data from the DIC system was used:

- Time t [s]
- Displacement of point 1, l_1 [mm]
- Displacement of point 2, l_2 [mm]
- To calculate the nominal stress and strain, it was used:

$$S = \frac{F_i}{A_0} \ [MPa] \tag{3.1}$$

$$e = \frac{\Delta l}{l_0} = \frac{l_1 - l_2}{l_0} \ [-] \tag{3.2}$$

Where the initial area of the cross-section, A_0 is given by,

$$A_0 = w_0 * t_0 \ [mm^2] \tag{3.3}$$

where l_0 and t_0 are the initial gauge length and width of the test specimen, respectively. All initial measurements were taken 3 times and an average was then computed and use for computing mechanical properties.

The determination of the true stress and true strain, was obtained by,

$$\sigma = \frac{F_i}{A_i} \ [MPa] \tag{3.4}$$

$$\varepsilon = \ln\left(\frac{l_i}{l_0}\right) = \ln\left(\frac{A_0}{A_i}\right) \ [-] \tag{3.5}$$

The relation between length and area on both equations 3.4. and 3.5. is based upon assuming incompressibility:

$$v_0 = v \Leftrightarrow A_0 * l_0 = A * l \Leftrightarrow \frac{l}{l_0} = \frac{A_0}{A} \tag{3.6}$$

Whereas it is known that plastics are compressible materials, equation 3.6. approximates it to incompressible behaviour.

Figure 3.5 shows different nominal stress - nominal strain evolution for plastics.

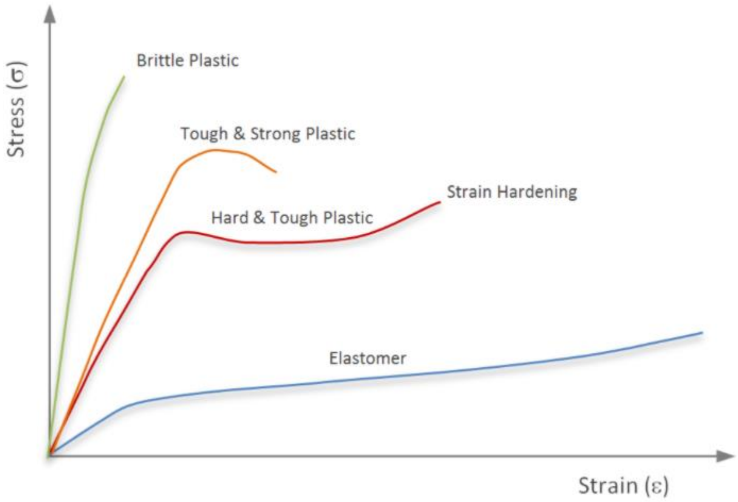


Figure 3.5. Different stress-strain curves for plastic materials (PolymerDatabase, n.d.)

It can be concluded that there are different possibilities for mechanical behavior of plastics. Not all plastics present a defined elastic region (linear) and some curves show that the specimens did not reach the necking stage (brittle plastic). Conversely, some curves show an almost total elastic region (elastomer).

The mechanical properties determined for all the tensile tests were the modulus of elasticity, E, the nominal yield strength, S_Y, the true yield strength, σ_Y, the nominal tensile strength, S_{ts}, the true tensile strength, σ_{ts}, the nominal rupture strength, S_r and the true rupture strength, σ_r.

The modulus of elasticity, E is calculated in any segment of the elastic and linear portion of the force vs displacement curve corresponding to the slope of this linear region. The yield point corresponds to the transition between elastic and plastic deformation and the determination of the nominal yield strength, S_Y and the true yield strength, σ_Y are obtained by equations 3.1 and 3.4, respectively for the yield point.

The instability point that corresponds to the onset of necking allows determining the nominal tensile strength, S_{ts} and the true tensile strength, σ_{ts} by means of equations 3.1 and 3.4.

Finally, the rupture point corresponds to the fracture of the specimens and allows determining the nominal rupture strength, S_r and the true rupture strength, σ_r by means of equations 3.1 and 3.4.

For the computation of results, a program in python was used to match the results from the INSTRON machine's system and the ones obtained from the DIC system. The program matched the initial instant picked up by the machine and the one from DIC system. It then matched the load registered by the INSTRON machine at which instability began and the picture taken by the DIC system at the same instant. By doing so, it was possible to match the registered load in the machine and the corresponding displacement of the gauge length in the specimens registered by the pictures and the DIC system.

3.2. Test specimens' manufacturing

There were four materials used to manufacture the different test specimens:

- Recycled PP (rPP)
- Virgin PP (vPP)
- Recycled HDPE (rHDPE)
- Virgin HDPE (vHDPE)

The properties of all materials, specified by suppliers, are detailed in Table 3.1.

Table 3.1. Properties of all materials used.

Material	Physical properties				Pellets' dimensions		
	Melt flow index [g/10 min]	Method (MFI)	Density [g/cm3]	Method (Density)	Average diameter [mm]	Average height [mm]	Color
vPP	45	ASTM D-1238-L	0,905	ASTM D-792A	4.2	1.1	White
rPP	2.0 – 4.5	ISO 1133-1	0,9 – 0,96	ISO83 - 1	4.5	1.3	Dark blue
vHDPE	0.5	ASTM D-1238-L	0.93 - 0.95	ASTM D-792A	5.2	2.1	White
rHDPE	0.4 – 0.8	ISO 1133-1	0,93 – 0,98	ISO83 - 1	4.6	2.7	Dark green

Although the supplier of the recycled material did not specify its origin, it did inform that although classified as ocean plastic waste, it is in fact picked up before in rivers and channels before reaching the ocean. This is relevant because the material did not spend as much time deteriorating as ocean solid plastic waste (SPW) found in the large concentration areas mentioned in section 2.1.5., which likely resulted in better properties.

Additionally, the make-up of the batches collected in these spots is likely to be more homogeneous than the one from open-waters SPW.

Figure 3.6 shows the different materials' pellets sizes and colors, and the geometry and average dimensions of each are presented in Table 3.1.

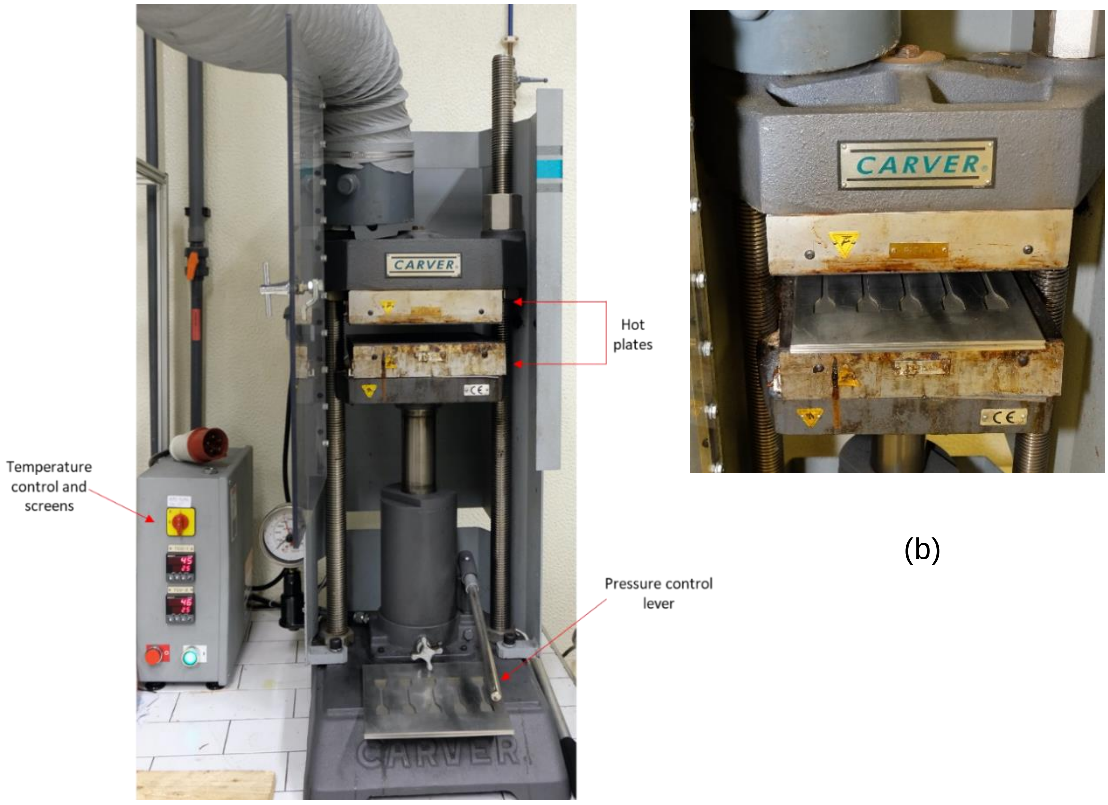
(a) (b) (c) (d)

Figure 3.6. Pellets of the different plastics used: rPP (a), rHDPE (b), vPP (c) and vHDPE (d)

All pellets, recycled and virgin, had a somewhat regular geometry but rather different sizes, which made more difficult the material flow inside the mold. The mold used to manufacture the test specimens is shown in Annex E. Figure 3.7 shows the difference in geometry and size of HDPE recycled and virgin pellets for a 50% recycled – 50% virgin mixture.

Figure 3.7. Difference in size and geometry of the rHDPE (blue) and vHDPE (white) pellets inside the mold

The test specimens were manufactured with a plastic heat press, model Carver M-2089 (Figure 3.8) in the Hands on Polymer Laboratory of IST-CERENA. The mold used allowed for the manufacturing of 5 specimens at a time. The mold geometry is available in Annex E.



(a)

(b)

Figure 3.8. The Carver M-2089 heat press (a) and detail of the empty mold between the hot plates (b)

The heat press used, the Carver M-2089, has a maximum of 250 kN of force and a range of 30ºC to 500ºC.

The manufacturing process consists of the following steps:

1. Mold coating with a demolding agent (silicone spray)
2. Pellets placement inside the mold (Figure 3.7)
3. Mold placement between the hot plates of the heat press
4. Heat press parameters definition, namely curing temperature, curing time, pressure
5. Cooling, firstly with a hair dryer and then by applying pressurized air to the closed mold
6. Opening of the mold
7. Removing the test specimens from the mold

The initial specimens produced allowed to verify the importance of the mold coating (step 1), the definition of an optimized curing temperature and time (step 4) and the definition of the cooling operation (step 5), Figure 3.9. presents some of the obtained results varying these parameters.

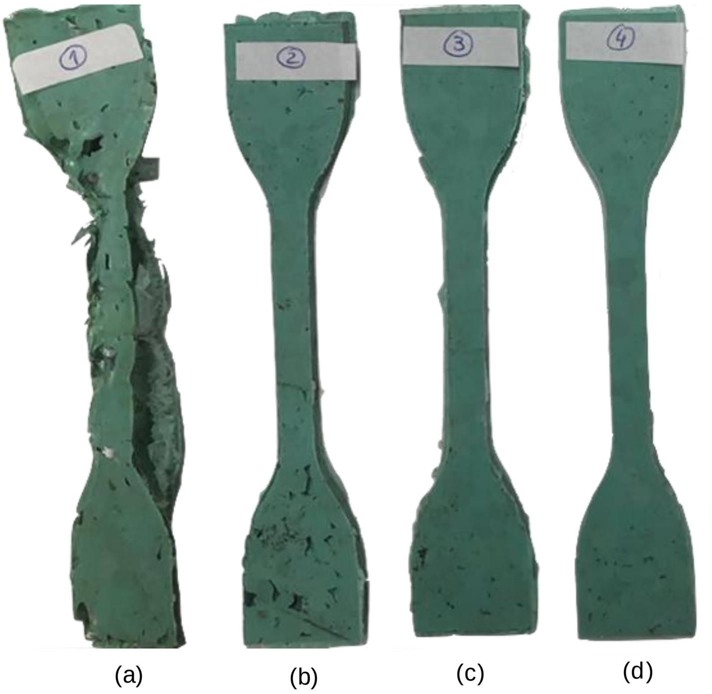


Figure 3.9. First four test specimens made: (a) using no demolding (b) curing for a too short of time (c) cooling at room temperature (d) and cooling with pressurized air

Aside from the initial experiments, there were two differences in the final test specimens made to try to improve their quality, namely by changing the temperature, curing time and applied pressure. Group 1 test specimens, referenced as M1, the pressure gradient was slowly applied increasing the force from 5 to 10 T during the curing time and additionally these were subjected to a cooling time of 15 min.

The test specimens in group 2, referenced as M2, the pressure was applied with no pressure gradient of a force of 8 T, and the cooling time was of 10 min.

The experimental plan in the next section details all the parameters used for the manufacturing of the test specimens.

3.3. Experimental plan

As mentioned before, to improve the quality of the test specimens, some preliminary tests were made, using different manufacturing methods: M1 and M2. Table 3.2. describes these tests.

Table 3.2. – Initial tests with different manufacturing methods M1 and M2

Reference	Material	Curing temperature [ºC]	Curing time [mins]	Pressure [T]	Cooling time [min]	Number of specimens
100% rPP_M1	rPP	190	10	5 to10	15	3
100% rPP_M2	rPP	190	10	8	10	3
75% rPP-25% vPP_M1	75% rPP – 25% vPP	190	10	5 to 10	15	3
75% rHDPE-25% vHDPE_M2	75% rHDPE – 25% vHDPE	160	10	5 to 10	15	3

The final experimental plan was defined as shown in Table 3., where different processing conditions and different percentage of recycled and virgin mixtures were considered. The material combinations considered specimens of PP and specimens of HDPE; the mixture of both materials was not considered.

Table 3.3. Experimental plan.

Reference	Material	Curing temperature [ºC]	Curing time [mins]	Pressure [T]	Cooling time [min]	Number of specimens
100% rPP_M1	rPP	190	10	5 to10	15	3
100% rPP_M2	rPP	190	10	8	10	3
100% vPP_M1	vPP	190	10	8	10	3
rPP - vPP_M1	100% rPP – 0% vPP	190	10	5 to 10	15	5 (of each)
	75% rPP – 25% vPP					
	50% rPP – 50% vPP					
	25% rPP – 75% vPP					
	0% rPP – 100% vPP					
rHDPE-vHDPE_M1	100% rHDPE – 0% vHDPE	160	10	8	10	5 (of each)
	75% rHDPE – 25% vHDPE					
	50% rHDPE – 50% vHDPE					
	25% rHDPE – 75% vHDPE					
	0% rHDPE – 100% vHDPE					

Figure 3.10 shows the mold filled with the different mixtures of HDPE.

Figure 3.10. Mold prefilled with mixtures of recycled and virgin HDPE: 100% rHDPE, 25% rHDPE, 50% rHDPE, 75% rHDPE and 0% rHDPE

As seen in the experimental plan and in Figure 3.10, different mixtures were processed at the same time using the same parameters. Although some testing was done to evaluate if different processing parameters would improve quality of different mixtures, time and material constraints made it impossible to further optimize the process. Even so, no significant difference was observed during testing of processing parameters.

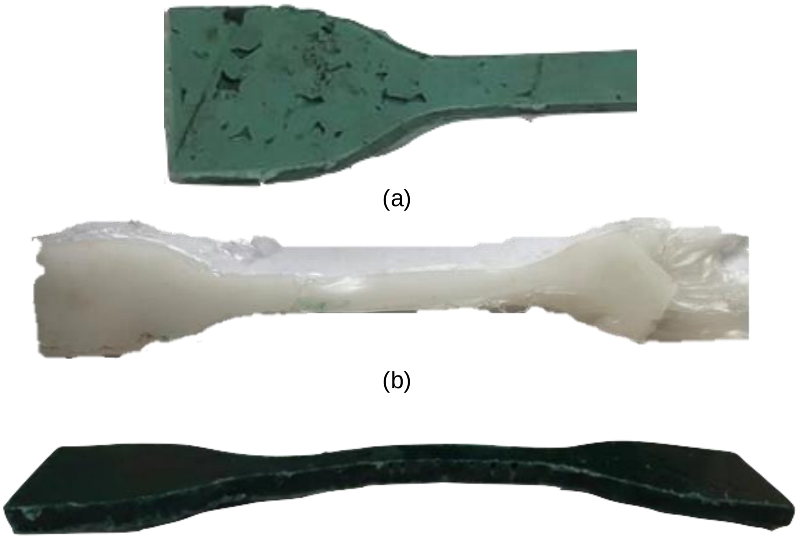
Considering the manufacturing and time constraints, it is worth noting the reason why special focus was put on the 75% recycled – 25% virgin material mixtures. After reviewing literature and visiting a factory producing fast-moving consumer goods (FMCG), it was identified that mixtures of about 80% recycled – 20% virgin material were often used because they guarantee high quality standards (equivalent to those found using 100% virgin material). A 75% recycled – 25% virgin material mixture would greatly decrease the amount of virgin material used. Naturally, “recycled” material in industrial applications is often of a much higher quality because it does not go through extensive degradation (it is usually made up of faulty parts that are shredded to use within the same facility). Even so, this mixture is especially significant because it can set a standard for industrial applications.

4. Results and discussion

This chapter begins by detailing the results of the manufacturing and preparation of the test specimens, where some difficulties occurred in the manufacturing methodology and due to the used materials. Afterwards, the different test results of the experimental plan were compared and will be shown.

4.1. Manufactured specimens

It is worth noting there were some limitations in the manufacturing of the test specimens. The defects caused by the limitations during the manufacture processing included superficial and inner gaps (Figure 4.1 (a) and (b)) pellets that did not melt properly (Figure 4.1 (b)), poor demolding (Figure 4.1 (c)) and warping of the test specimens (Figure 4.1 (d)) as well as localized burning of the pellets.

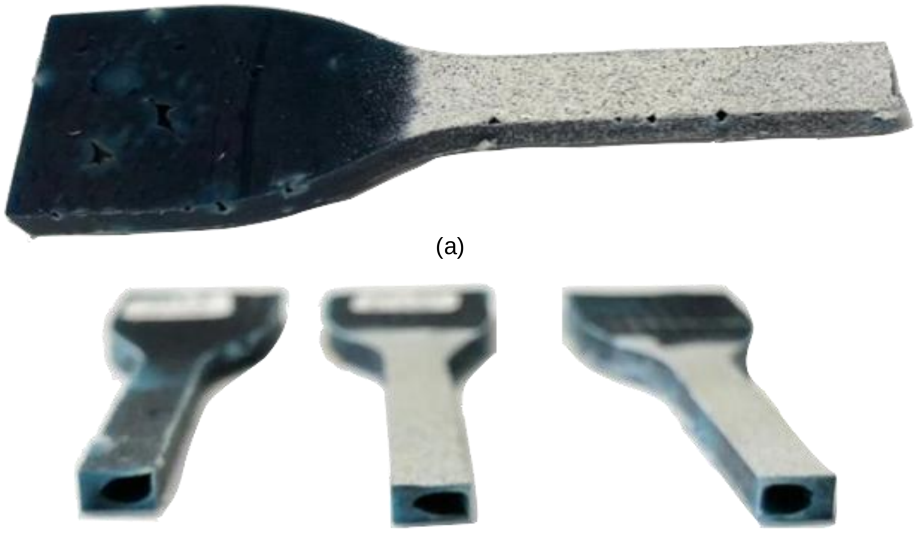

(a)

(b)

(c)

Figure 4.1. Test specimens' defects: (a) superficial material gaps in one specimen of rPP, showing pellets that did not melt during curing process (b) poor demolding and consequent deformation in vPP specimens (c) warping

One of the most significant defects found in the specimens were the material gaps. Figure 4.2 (a) shows a detail of three rPP mixtures' test specimens that have fractured in a zone with a significant material gap.



(a)

(b)

Figure 4.2. Fracture for three 75% rPP mixture test specimens: (a) gaps in the gauge length (b) closer detail of the fracture area

These specimens had not only inner gaps but also superficial ones (Figure 4.2 (b)). This issue made the mechanical behavior of these specimens very fragile and they presented practically no ductile behavior.

As stated in chapter 3, the two manufacturing conditions (M1 and M2) were defined to improve the quality of the test specimens, however, due to the occurrence of the inner gaps these revealed no significant differences.

Some alternative manufacturing conditions were additionally tested in order to overcome the inner gaps defect as:

- Filling the mold with more pellets than needed (overfilling the mold);
- Turning the mold halfway through the curing time;
- Different cooling conditions using a hairdryer, pressurized air or leaving the mold to cool down at room temperature;
- Variation of the cooling time.

The overfilling of the mold technique allowed to improve one of the specimens' surface, however, it did not alter the quality of the test specimens, Figure 4.3 shows the result after demolding one of the plates with specimens of 100% rPP.

Figure 4.3. Speciments of 100% rPP when the mold was overfilled.

Superficial gaps can be seen on the surface of the specimens. Additionally, there was a was always an issue with the last plate to be removed from the mold. One of the sides of the specimens would always present a much worse surface than the other. Figure 4.4. shows two different sides of the same two test specimens.

(a) (b)

Figure 4.4. (a) Front side (b) back side of two test specimens (first one made of a mixture of PP and second one made of a mixture of HDPE)

While the front side presents a clean, smooth surface, the back side of the specimens shows gaps and poor demolding. Regardless of which side the plate was facing (upper part of heat press or lower one) this issue always happened.

Although manufacturing conditions were changed, the plastic considered or the mixture used, the inner gaps defect always occurred. Some of the reasons why this issue occurred can be the following:

- Polymer incompatibility – regardless of the process used, it is important to ensure that the polymer mixtures (recycled and virgin) are suited to the process/equipment used. Since the MFI of the recycled and virgin polymers was very different in the case of PP, this significantly hindered the results of these specimens;
- Hot-pressing limitations – the manufacturing process is extremely fast in producing test specimens for a relatively low cost, when compared with extrusion and injection processes, and with minimized distortion. However, it is very hard to control the processing parameters for hot-pressing. Not only is the variation in the applied pressure is difficult to control, since it is done manually, but the temperature can vary drastically between the set temperature, the one reached once the press is opened and the “cold” mold is put inside, and the one once the curing time is completed;
- Cooling procedure - once the mold is taken outside it needs to be induced the cooling, with a hairdryer and pressurized air, otherwise the process would be significantly time-consuming. This method is performed manually and lacks proper control, which is further highlighted by the complex geometry of the specimens.

An alternative manufacturing process for the tensile specimens would be extrusion, followed by injection. The need to manufacture tensile specimens with different mixtures, it would require to deep clean of the extrusion equipment to be thoroughly cleaned after being used and not contaminate the mixtures. This requirement would increase significantly the material needed and the time to manufacture all the combinations defined in the experimental plan. Even so, a test was done with a mixture of 75% recycled and 25% virgin PP where a small extrusion machine was used. Figure 4.5. shows the material produced and the one of the specimens produced.

(a) (b)

Figure 4.5. (a) Pellets of the 75% recycled – 25% virgin PP extruded (b) detail of a specimen of the same mixture tested with an uneven surface

This specimen did not present significantly higher quality than the ones produced without using the extrusion machine.

Since the amount of recycled material available was limited, extrusion was not an option because it requires significative and continuous input of material. Even considering the material availability and time limitations, a manufacturing test was performed in a small extrusion machine to test if the polymers (recycled and virgin) would melt better once previously mixed. After extruding the material, the pellets were then again cured in the same heat press used to produce the other specimens. No visible differences were verified in these test specimens.

Even though the experimental plan was to test 5 specimens for each type of mixture, these did not present the required quality to be tested. For this reason, in some cases only 3 test specimens could be tested and in others (namely in the different percentages' mixtures) only 1. The best test specimens were chosen in regard to their physical appearance. Test specimens were chosen by visually inspecting the gauge length for porosity.

4.2. Virgin and recycled polypropylene

For the comparison of 100% virgin and 100% recycled polypropylene (PP) specimens, three test specimens of each group were tested. To investigate the specimens' quality, two different manufacturing conditions (M1 and M2) were considered.

As detailed in the experimental plan, test specimens in M1 were done with slow increases of pressure, reaching a higher maximum pressure, and Figure 4.6. shows the load-displacement plot for the best test specimens of each condition.

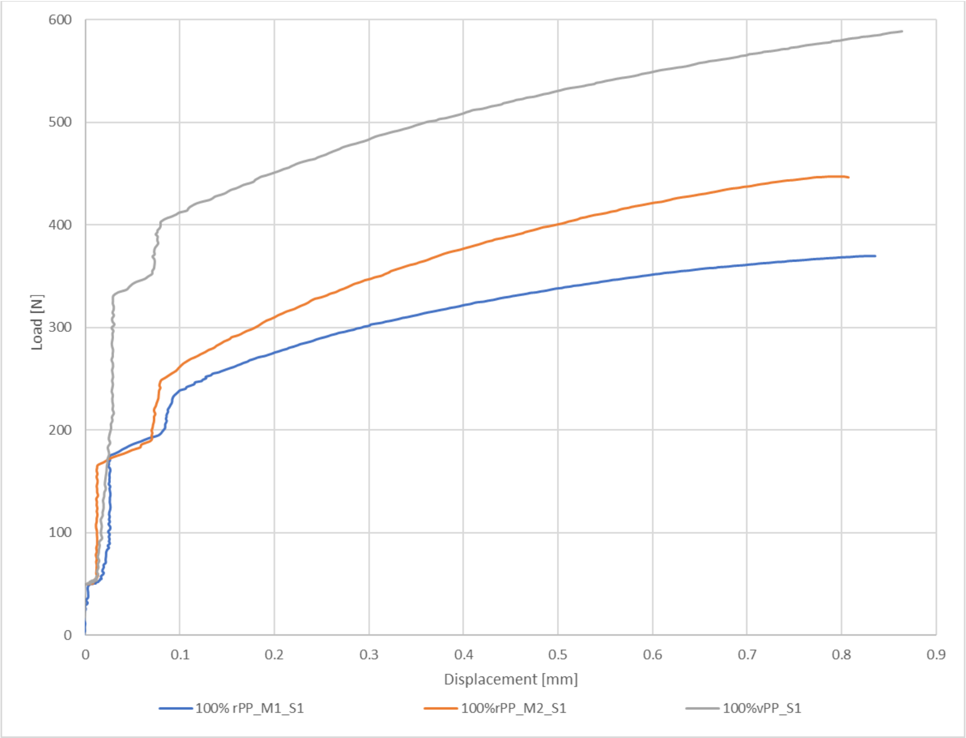
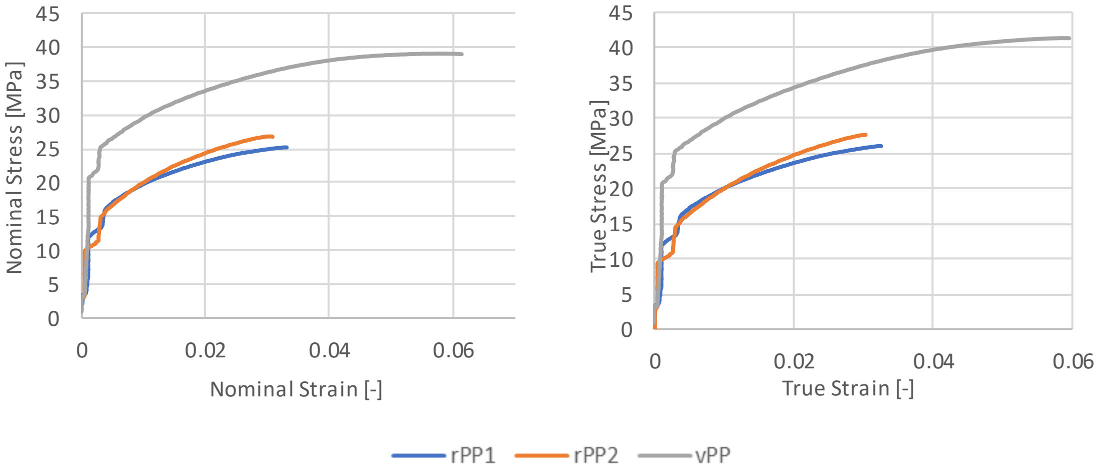


Figure 4.6. Load – displacement curve for 3 specimens of 100% recycled and 100% virgin PP

The initial instability in the plots might be due to the compatibilization of data from the INSTRON machine and the DIC system. The comparison of the two best test specimens of the two recycled specimens (100%rPP_M1 and 100%rPP_M2), it can be concluded that the difference in the manufacturing parameters did not improve the quality of the test specimens. On the other hand, it is worth noting that for all specimens the beginning of the load-displacement curve revealed some irregularities than can be associated with the inner gaps inside the material.

Additionally, it can be concluded that the 100% virgin PP test specimens have a better mechanical behavior than the recycled specimens, as it was expected. Figure 4. shows the nominal stress-strain curve and the true stress-strain curve for the three specimens.



(a) (b)

Figure 4.7. Nominal stress – nominal strain (a) and true stress – true strain (b) curves for 3 specimens of 100% recycled and 100% virgin PP

Table 4.1 shows the mechanical properties of the three test specimens.

Table 4.1 . Mechanical properties for 3 specimens of 100% recycled and 100% virgin PP

Specimen reference	Nominal yield strength, $S_Y\ [MPa]$	True yield strength, $\sigma_Y\ [MPa]$	Modulus of Elasticity, $E\ [GPa]$	Tensile strength nominal, $S_{ts}\ [MPa]$	Tensile strength true, $\sigma_{ts}\ [MPa]$	Nominal strain at brake, $A\ [\%]$
100% rPP_M1_S1	15.82	15.50	4.3	25.17	26.00	3.3
100% rPP_M2_S1	14.89	15.91	3.9	26.79	27.60	3.1
100%vPP_M1_S 1	24.97	24.29	7.4	39.02	41.33	6.1

It can be concluded that the yield strength values are within the reasonable interval for PP (10 - 500 MPa) and that the 100% recycled specimens have somewhat similar mechanical properties, aside from the Modulus of Elasticity. It is worth noting that this parameter was obtained from the tensile testing, which is not the recommended way to do it (through cycles of load). Because there is such variability of mechanical behaviour of polymers (Figure 3.5), the method to compute the Modulus of Elasticity depends on the stress – strain curves of the material analyzed.

Because the test specimens had such small linear regions (due to the inner gaps of material in the gauge length) this value is likely not representative of the true mechanical behavior of the material. The 100% virgin test specimen has an overall better mechanical behavior and was able to produce a much longer nominal strain at brake.

Aside from the manufacturing issues, it would be expected that the recycled and virgin materials would have significantly different properties due to their other characteristics.

4.3. Mixtures of virgin and recycled polypropylene

As mentioned before, the poor quality of the test specimens made it so that only one test specimen of each mixture could be tested. Figure 4. shows the nominal stress-strain curve and the true stress-strain curve for the five specimens shows the load-displacement plot for the best test specimens of each type of mixture, from 100% recycled PP (rPP) to 100% virgin PP (vPP). All these specimens were made under the manufacturing condition M1.

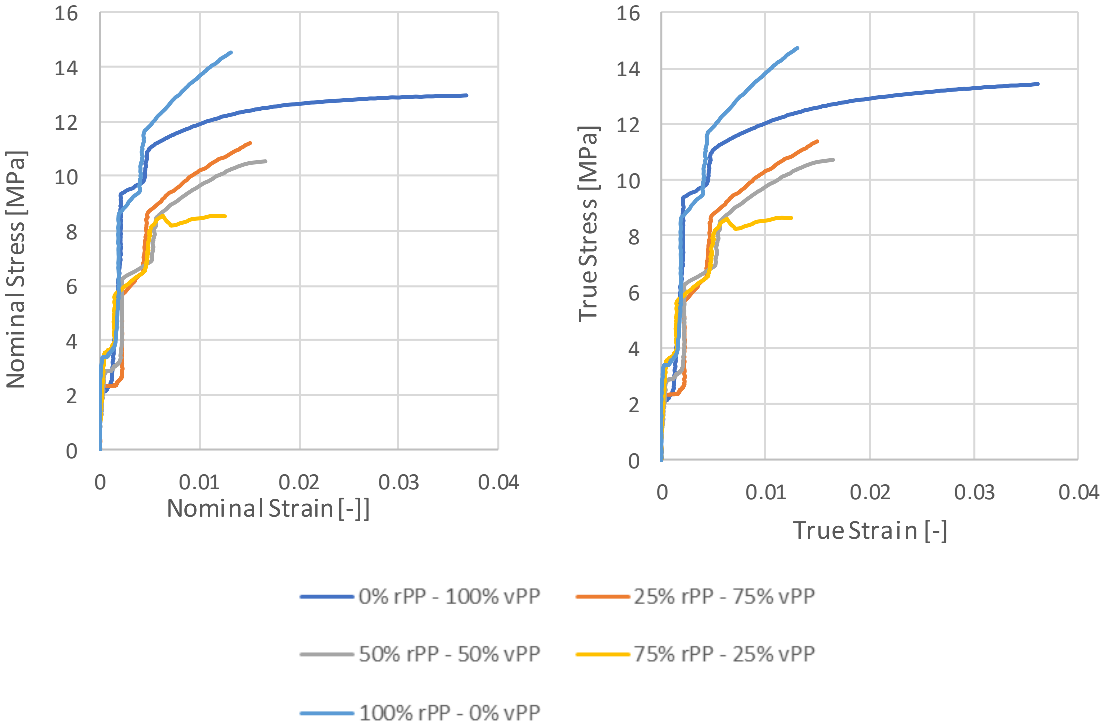


Figure 4.8. True stress – true strain (a) and nominal stress – nominal strain (b) curves for 5 specimens of 100% recycled and 100% virgin PP

From Figure 4. it is visible that the mechanical behavior of the test specimens is not cohesive. This can be due to issues mentioned before, such as the manufacturing process. No significant conclusion can be drawn from Figure 4.. as the two test specimens with the best mechanical behavior are the ones made of 100% recycled and 100% virgin PP. This fact supports the hypothesis that both polymers did not flow together well, making the mixtures more unsuccessful than the 100% test specimens.

Table 4.2 shows the mechanical properties of the 5 test specimens.

Table 4.2. Mechanical properties for 5 test specimens with different percentages of rPP.

Specimen reference	Yield strength nominal, $S_Y\,[MPa]$	True yield strength, $\sigma_Y\ [MPa]$	Modulus of Elasticity, $E\ [GPa]$	Tensile strength nominal, $S_{ts}\,[MPa]$	Tensile strength true, $\sigma_{ts}\ [MPa]$	Nominal strain at brake, A [%]
0% rPP	10.84	5.94	2.04	13.00	7.18	3.7
25% rPP	8.49	8.80	1.47	11.21	11.38	1.5
50% rPP	8.47	8.43	1.35	10.54	10.72	1.7
75% rPP	8.1	7.88	1.38	8.55	8.68	1.6
100% rPP	11.65	11.58	2.07	14.52	14.72	1.3

From Table 4.2 no significant conclusions can be draw. Although the 100% virgin PP specimen has the highest nominal strain at brake, the 100% recycled specimen presents the best mechanical properties overall. The mixtures present overall worse properties than the 100% recycled and virgin specimens. It is worth noting that all the mixtures (25%, 50% and 75% recycled PP) have very similar properties, making an interesting point to the possibility to increase the amounts of recycled plastic in industrial applications without significant loss of properties.

4.4. Mixtures of 75% recycled – 25% virgin polypropylene

As mentioned before, special focus was put into the 75% recycled – 25% virgin mixtures since they are commonly used in the industry to reduce costs and material waste. Although it was possible to test 3 specimens, only 2 were able to produce results in the DIC software. This issue may have happened because of the uneven surface of the specimens – due to the issue of poor quality of the surface, the paint of the stochastic pattern was not successfully achieved for all of specimens. For this reason, the degree of accuracy in the software used to compute displacements had to be decreased (for the program to pick up the pattern).

Figure 4. shows the nominal stress-strain curve and the true stress-strain curve for the 2 test specimens of a 75% recycled - 25% virgin mixture of PP.

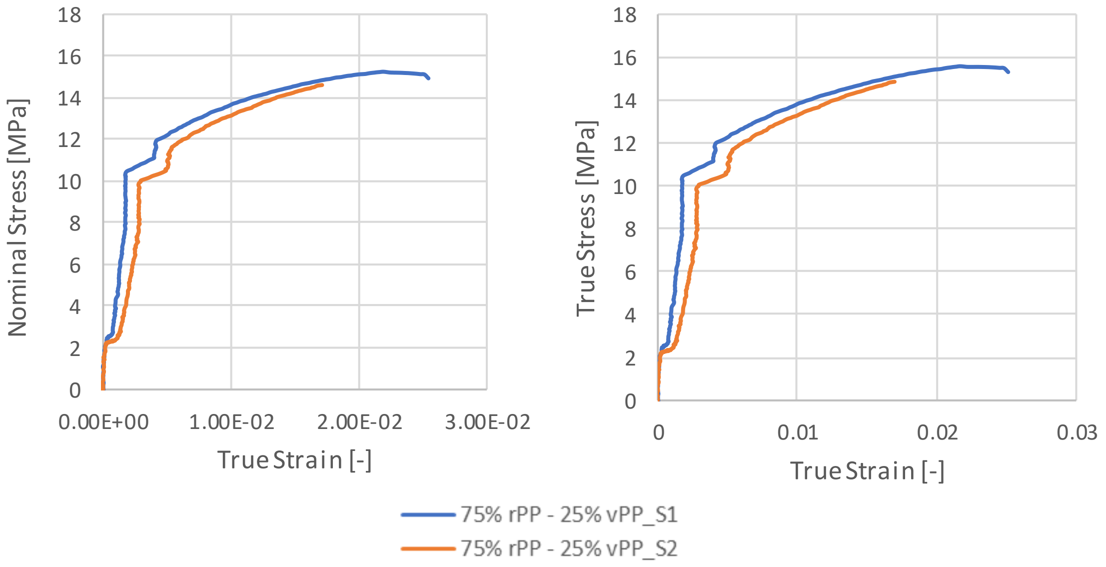


Figure 4.9. True stress – true strain (a) and nominal stress – nominal strain (b) curves for 2 specimens of 75% recycled and 25% virgin PP

It can be concluded that both specimens have a somewhat similar mechanical behavior, which shows some reproducibility of the results. Consequently, this reveals some consistency on the appearance of inner gaps in the material. Table 4.3 shows the mechanical properties of the 2 test specimens, (specimen 1 – S1 and specimen 2 – S2).

Table 4.3. Mechanical properties for 2 test specimens of a 75% recycled - 25% virgin mixture of rPP

Specimen Reference	Yield strength nominal, $S_Y\ [MPa]$	Yield strength true, $\sigma_Y\ [MPa]$	Modulus of Elasticity, $E\ [GPa]$	Tensile strength nominal, $S_{ts}\ [MPa]$	Tensile strength true, $\sigma_{ts}\ [MPa]$	Nominal strain at brake, $A\ [\%]$
75%rPP-25%vPP_S1	11.93	11.98	2.54	15.25	15.58	2.5
75%rPP-25%vPP_S2	11.48	11.17	1.95	14.67	14.87	1.7

Table shows that both specimens have similar mechanical properties and that the test specimen S1 performs better overall.

4.5. Mixtures of high-density polyethylene

The HDPE specimens often presented problems in the computation of results. For the 75% recycled – 25% virgin HDPE the DIC software was not able to compute data, problably due to issues of uneven surface. Figure 4. shows the nominal stress-strain curve and the true stress-strain curve for the 4 specimens of mixtures of recycled and virgin HDPE.

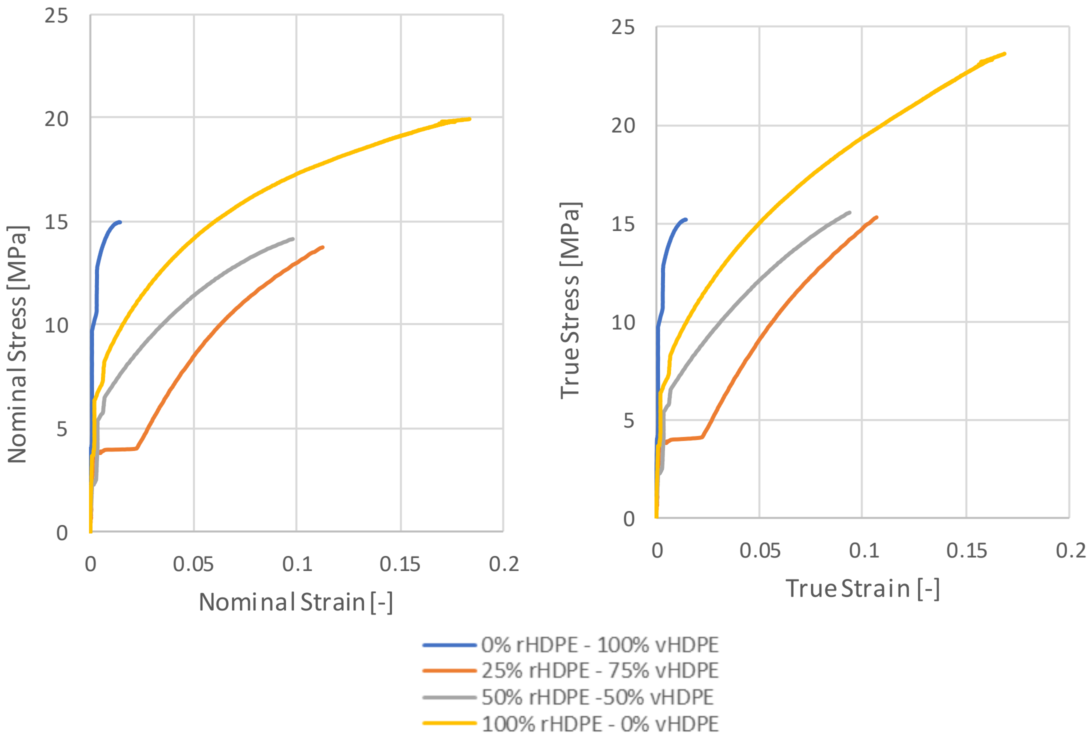


Figure 4.10. True stress – true strain (a) and nominal stress – nominal strain (b) curves for 2 specimens of 75% recycled and 25% virgin PP

These curves are incomplete as the DIC system could not compute all the data from the test (the test continued for all curves except for the 100% rHDPE – 0% vHDPE specimen), therefore these results are not an accurate depiction of mechanical behaviour of the specimens. It can be seen there is a great deal of dispersion in the HDPE specimens' results. It is worth noting that the data for force for the 25% rHDPE – 75% vHDPE, 50% rHDPE – 50% vHDPE and 0% rHDPE – 100% vHDPE, continues beyond the curve shown but no displacement was possible to compute. For this reason, it can be assumed that the specimen with the best mechanical behaviour would be the 100% virgin HDPE, which was to be expected. On the other hand, the 100% recycled HDPE specimen shows a better mechanical performance than the mixtures. As for the PP specimens, this is likely due to the deffects in the gauge length and difficult compatibilization of the virgin and recycled polymers.

Table 4.4 shows the mechanical properties of the 4 test specimens.

Table 4.4. Mechanical properties for 2 test specimens of a 75% recycled - 25% virgin mixture of PP

Specimen reference	Yield strength nominal, $S_Y\ [MPa]$	Yield strength true, $\sigma_Y\ [MPa]$	Modulus of Elasticity, $E\ [GPa]$	Tensile strength nominal, $S_{ts}\ [MPa]$	Nominal strain at brake, A [%]
0% rHDPE	9.71	13.20	3.13	15.00	1.4
25% rHDPE	3.77	4.06	2.56	13.75	11.5
50% rHDPE	5.35	6.40	0.87	14.14	9.8
100% rHDPE	6.21	8.44	1.08	19.95	18.4

It can be concluded that the 100% virgin specimen performs better overall and that the 100% recycled specimen has a considerably higher nominal strain at brake than all the other specimens. Once again, the mixtures proved to have worst mechanical properties than the 100% recycled and pure specimens, supporting the hypothesis that proper compatibilization did not occur.

4.6. Mixtures of 75% recycled – 25% virgin high-density polyethylene

As mentioned before, several issues prevented the proper testing and processing of results of the test specimens. In this case, only 2 test specimens were possible to test. Quality issues made it impossible for the DIC software to compute any results for all the specimens tested. The reason this issue happened was because there were superficial gaps in the test specimen. The gaps make for an uneven surface and make it impossible for the DIC system to pick up the pattern. Figure 4.5 shows one of the specimens tested where the superficial gaps can be seen.

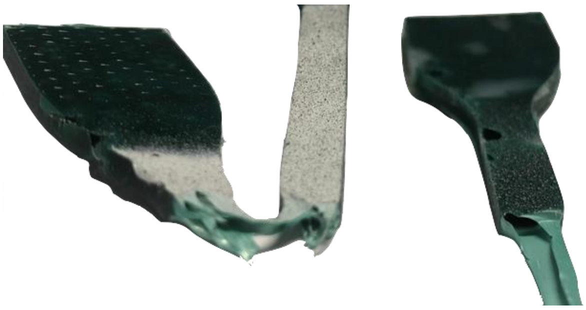


Figure 4.5. 75% recycled – 25% virgin HDPE test specimen that did not reach fracture

To analyze the specimens, the load – displacement plot was computed using load data from the testing machine and the displacement data from the grips (tensile testing machine). Figure 4.6 shows the load – displacement curves for the specimens.

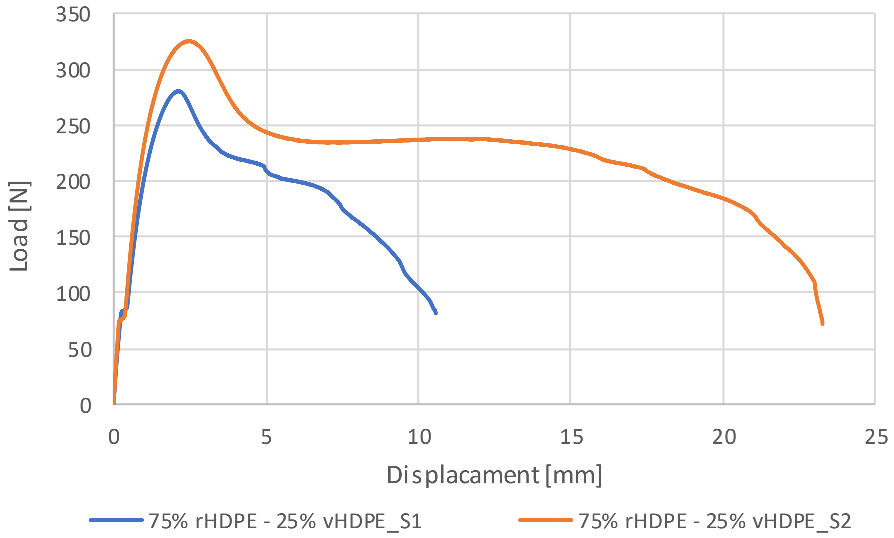


Figure 4.6. Load – displacement curve for 2 test specimens of a 75% recycled - 25% virgin mixture of HDPE

The test specimens show a somewhat different mechanical behavior, with the second specimen presenting a much better mechanical performance. Once again, the HDPE specimens show a lot of dispersion in the results, making it difficult to draw significant conclusions.

5. Conclusion and future work

To better understand the characteristics and impact of solid plastic waste in the ocean, a review of literature on this issue was done. To mitigate this problem, Circular Economy strategies were applied to reach the conclusion that the use of mechanically recycled plastic could present a solution. To maximize the use of recycled plastic, this work aimed to develop a simple methodology to perform mechanical characterization of mixtures of unknown recycled ocean plastic and virgin plastic. This characterization was done through tensile testing.

Although the manufacturing of the test specimens hindered the results that could be obtained from the methodology, it also highlighted the importance of using a high-standard manufacturing process to get accurate results. An appropriate manufacturing process will allow for detail in comparing virgin and recycled materials and for more reliability in increasing amounts of recycled percentages in plastic products without significant loss of the materials' or parts' properties. The manufacturing of the tensile test specimens should likely be done by extrusion and injection.

It was still possible to identify that even recovered materials of poor quality, which have been extensively degraded by exposure to elements in the ocean, present similar mechanical behavior of their virgin counterparts and can demonstrate reasonably good properties. Additionally, it was found that due to poor compatibilization of the recycled and virgin plastics, mixtures of both presented overall poorer mechanical properties than the 100% recycled or virgin test specimens.

The level of quality found in the recovered materials and the mixtures observed make them suitable for widespread use in industrial applications and the FMCG market, where plastic is widely used.

In the future, appropriate manufacturing and virgin materials should be used to replicate different-percentage mixtures and evaluate their mechanical properties and how well they compare to the virgin counterparts.

A suitable manufacturing method for the test specimens includes extrusion followed by injections molding. Additionally, the polymer pellets should be baked out before being processed to remove impurities and residual humidity.

Moreover, additional mechanical testing should be performed for 75% recycled – 25% virgin mixtures because this percentage seems to have already been accepted empirically in industrial applications as a “safe” one to use.

Finally, applications for the different mixtures should be identified in an effort to streamline the use of recovered plastics, and channel the use of virgin materials to food-protection applications where their replacement is more difficult.

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Annex A

Synthesis of current knowledge, concerns and recommendations relevant to environmental and human health issues from production and plastic waste management (Thompson et al., 2009)

	Established knowledge	**Concerns and uncertainty**	**Recommendations for industry, research and policy**
Production and use	plastics are inexpensive, lightweight, versatile, water resistant and durable annual growth in plastic production is approximately 9% (currently >260 Mt yr^-1) around 8% of world oil production is used to make plastics plastics bring extensive societal, human health and environmental benefits >33% of production is used for disposable items of packaging	is our usage of hydrocarbons for plastics sustainable? to what extent could biopolymers replace oil-based plastics? is there enough arable land for production of biomass (crops) required for biopolymers? to what extent does use of plastic powders as cleaning abrasives, and scrubbers results in direct release of particles to environment?	increase/incentivize material reduction and reuse construct life cycle analysis of production, disposal/recycling of major polymers (including biopolymers, degradable and biodegradable polymers) and plastic products develop alternative monomers, polymers and additives using green chemistry approaches revise international standards for and introduce accurate/ informative labelling of recyclable, ‘degradable’, ‘biodegradable’ and compostable polymers

Disposal: waste management	Plastics are a substantial part of domestic and industrial wastes in landfill recycling of some polymers (e.g. PET) has increased considerably in recent years, but substantial quantities of plastic waste not compatible with recycling biodegradable polymers typically require industrial composting and will not readily degrade in landfill biodegradable plastics can compromise recycling	are current disposal strategies sustainable - lack of space in landfill? to what extent do chemicals leach from plastic in landfill? little is known about the degradability or environmental fate of additives used in biodegradable polymers	increase/incentivize product design towards use of recycled feedstock and increased end-of-life recyclability improve methods to collect and separate plastic waste for recycling investment in/incentivize recycling operations standardize labelling so consumers can identify products with high end-of-life recyclability (traffic light system) research and monitoring of leachates from landfills
Disposal: littering and dumping	plastic debris is common in marine habitats worldwide, including poles and deep sea the abundance of plastic debris is increasing/stabilizing (not declining) plastic debris is fragmenting, with pieces <20 μm on shorelines and in water column	to what extent will breakdown of plastic debris increase the abundance of small fragments in the environment? rates of accumulation of debris on land, in freshwaters and in the deep sea are not certain do biodegradable or compostable plastics degrade in natural habitats?	education/incentives to promote the value of end of-life plastics as a feedstock for recycling education and associated enforcement on the wasteful and adverse ecological effects of plastic spillage, dumping and littering develop standard protocols and monitoring to evaluate trends in the abundance of plastic debris across in natural habitats cleaning programmes in natural, urban, and industrial locations research on breakdown of degradable and biodegradables
Issues relating to wildlife	>260 species are known to ingest or become entangled in plastic debris ingestion is widespread in some populations (>95% of individuals) and can compromise feeding entanglement in plastic debris can lead to severe injury and death	does ingestion of, or entanglement in, plastic debris have effects at the population level, or can such effects combine with other stressors to do so? to what extent do plastics transport/release	research to establish the distribution, abundance and environmental consequences of micro- and nano-plastic fragments research to establish potential for plastics to transport chemicals to food chain research to establish population level consequences of ingestion and entanglement

		chemicals to wildlife? what are the consequences of the accumulation of small plastic particles (e.g. abrasives from cleaning applications) in the environment?	education, monitoring and cleaning

Annex B

Drivers of Circular Economy, adapted from Hopewell et al., 2009, David, 2017, Ragaert et al., 2017, Baytekin et al., 2013, Singh et al., 2017, J. Maris et al., 2018

Drivers of CE	Capital	Policy	Public Awareness	Technology	CE framework	Non-consensual definition	Non-universal standards and metrics	Health and environmental concerns
Govindan and Hasanagic (2018)	X	X	X	X	X			X
Mishra et al. (2018)	X	X		X				X
Ritzen and Sandstrom (2017)	X	X	X	X				
Elia et al. (2017)							X	
Torstensson (2016)	X		X	X				
Lieder and Rashid (2016)								
Rizos et al. (2016)								
Eijk (2015)								
Haas et al. (2015)						X		

Liu and Bai (2014)	X		X					
Su et al. (2013)	X	X						
Preston (2012)								
Geng and Doberstein (2008)	X	X	X	X				
Berchicci and Bodewes (2005)			X					

Annex C

Overview of mechanical recycling improvement/valorization techniques, adapted from Gu et al., 2017

Method	Description	Sources
Filler addition	Fillers can both improve properties of recycled plastics as well as lower production costs. Chemical families of fillers include inorganics – oxides, hydroxides, salt, silicates, and metals – and organics – carbon and graphite, and natural and synthetic polymers (Xanthos, 2003).	Dehghani et al., 2013; Al-Maadeed et al., 2014; Gu et al., 2016a; Sommerhuber et al., 2016 Sommerhuber et al., 2015
Compatibilization	Because most polymers are immiscible, compatibilization is the technique by which interfacial tension and stress interface are reduced to promote compatibility. This is achieved with chemical (e.g. copolymer) or physical (e.g. mineral nanoparticles) additives. Another use of compatibilization is when organic fillers are used as reinforcement for recovered plastics. Poor compatibility and low rates of moisture absorption in the resulting composites can be solved by adding reactive groups like maleic anhydride and acrylic acid.	Vazquez and Barbosa, 2016; J. Maris et al., 2017 Vilaplana et al., 2008 Ragaert et al., 2017
Process optimization	Due to injection parameters having a significant importance on the performance of plastic products, be them of virgin or recycled ones, one way to increase quality is to optimize said parameters. This analysis can be done by using the Taguchi experimental design method, (in a way to decrease number of experiments). In a study by Gu et al., (2014), this technique was found to not only find optimal levels of processing parameters but also to be a way of reducing number of trials and quickly identify the effects of processing parameters in the quality of end materials.	Gu et al., 2014; Altan, 2010; Fei et al., 2013; Mizamzul et al., 2011; Masato et al., 2017

Formula Design	In order to meet high-end industrial technical requirements, a special formula of the recovered plastic can be designed. A fairly recent technique, formula design is essentially used before other improvement techniques either through numerical modelling of the effects of each component (Homkhiew et al., 2014), composite design methods (Stark and Matuana, 2003, Soury et al., 2009, Zolfahgary et al., 2013) and lastly via a combination of hierarchical clustering analysis (HCA) and principal component estimate (PCE), (Gu et al., 2016)	Homkhiew et al., 2014; Gu et al., 2016b; Stark and Matuana, 2003; Soury et al., 2009; Zolfahgary et al., 2013
Virgin plastic mixture	Blending virgin polymers with recovered ones – not necessarily recycled but industrial scrap – is a widely used technique in the industry and often combined with the addition of stabilizers and fillers.	Sommerhuber et al., 2015 Vilaplana et al., 2008
Re-stabilization	Because degradation impacts mechanical properties and makes the material more susceptible to further deterioration, re-stabilization is a technique performed by adding stabilizers like hindered phenols and hindered amine stabilizers. These fillers do not “recover” the polymer but prevent it from suffering further degradation.	Vilaplana et al., 2008

Annex D

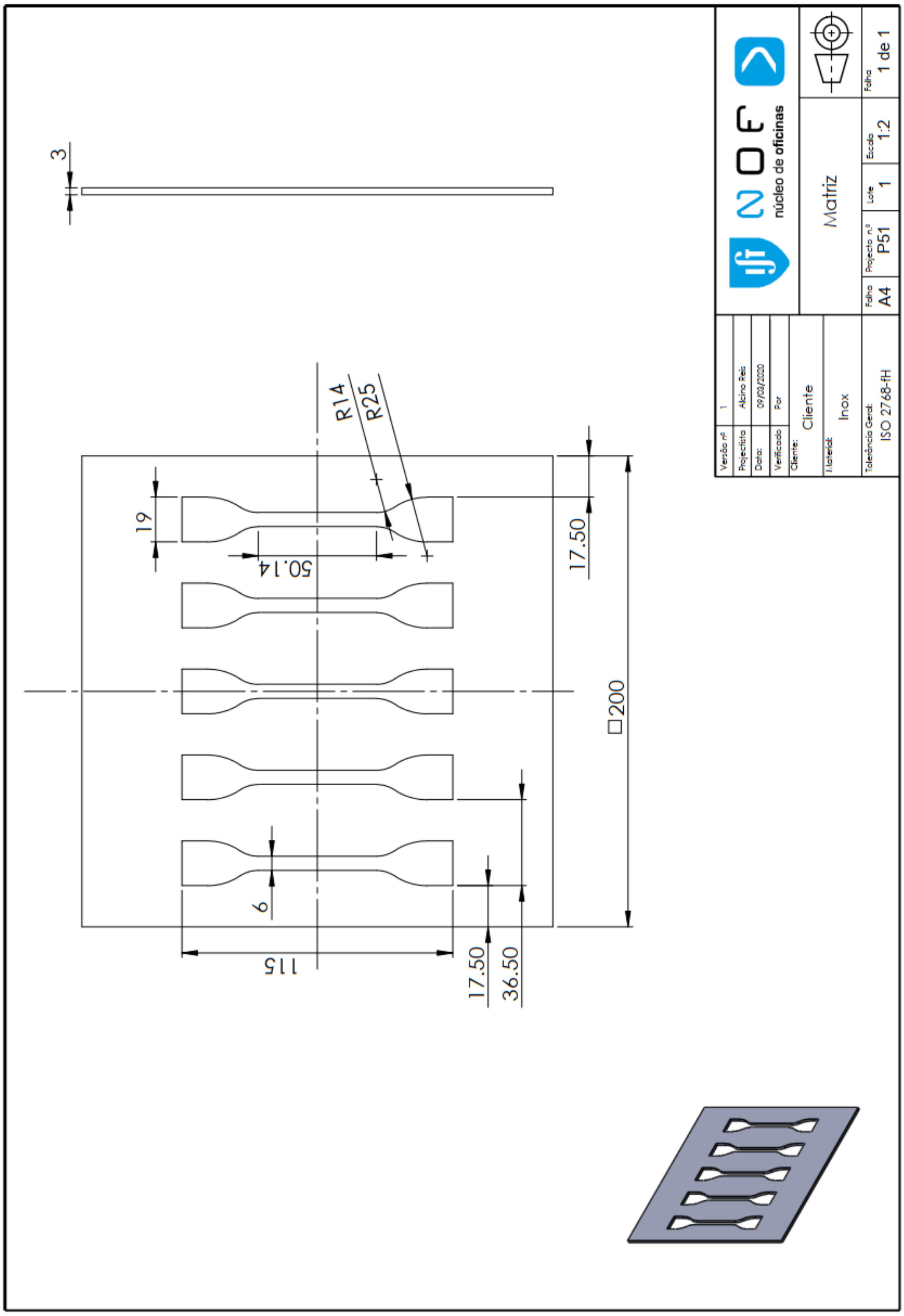
Summary of different case studies on performance of different recycled polymers.

Source of solid plastic waste (SPW)	Polymer	Application	Description	Comments and results	Source
Milk bottles	HDPE	Wood-plastic composites	Mechanical characterization through tensile, flexural and Charpy impact tests of different composition wood-plastic composites, using different sourced amounts of pure and recovered PE and wood waste.	Inorganic impurities and moisture were two factors which negatively impacted properties of test specimens. Virgin and recovered materials showed equivalent mechanical properties and water content up to a level of 60% of wood in the composite.	Sommerhuber et al., 2015
Unknown source, Turkey	LDPE, HDPE, PP	Unknown	Tensile tests of different composition mixtures of recovered and virgin single polymer specimens.	Mechanical properties were lower when compared with the 100% virgin equivalents, but it was possible to have a very small decrease of tensile strength in all polymers (15% for LDPE, 5% in HDPE and 3% in PP). PP was found to be the polymer with highest potential for mechanical recycling, but all materials were found to have a usability rate of 100%.	Meran et al., 2008
MSW, Ecocentral Granada, Spain	HDPE, PET, LDPE and HDPE film, PS+EPS	Unknown	Tensile properties (creep stress, deformation at creep point, resilience, deformation at resilience point, nominal deformation at breaking point), tear resistance, moisture content, MFI and impurities' analysis were performed in 100% recycled material and a mixture of 75%-25%.	Results obtained were that both mixtures present good properties, close to those of virgin PE, with possible manufacturing through blowing or flat films and injection molding.	Soto et al., 2018
Industrial and MSW, Denmark	PET, PE, and PP	Unknown	Thermal degradation, moisture content and mechanical properties (impact strength, tensile strain, and tensile strength) analysis were performed in a variety of reprocessed packaging including food-protecting.	Pet was found to be, in theory, suited for closed-loop recycling because potential degradation could be reversed in a decontamination process. Substantial differences were found for PET, PE and PP recycling, noting the importance of the need for specific design of recycling systems for each polymer.	Eriksen et al., 2019

Recycling plant, New Zealand	HDPE	Wood-plastic composites	Several mixtures with varying degrees of different grades of both virgin and recycled HDPE and wood flour were analyzed for density, moisture absorption, thickness swelling and tensile and flexural properties (strength, Young Modulus, yield stress, fracture, and maximum strain)	Composite panels made of recycled HDPE were found to be similar or to have better mechanical properties when compared to the ones made of virgin HDPE. Dimensional stability and high-quality mechanical properties (similar or better than the ones currently achieved with virgin polymers) can be attained by increasing polymer content or by adding coupling agents.	Adhikary et al., 2008
Reprocessed and recycled HDPE from sewer pipes' production, France	HDPE	Unknown	Mechanical testing, oxidative induction time (OIT), MFI and thermal analysis were used in conjunction with X-ray fluorescence (m-XRF), size exclusion chromatography and 13C solid-state NMR to evaluate mechanical behavior (Young's modulus (MPa) Yield strength (MPa) Strain at break (%) Strain at ultimate stress) and molecular structure as well as contaminant or filler contents of virgin and three types of recycled HDPE (with different contents of virgin polymers and different sourced plastic)	Even though recycled materials were found to have good quality, contaminants and filler residues were found to have considerable effects on properties of recovered polymers. To improve performance of recycled HDPE it is suggested to reduce the amount of contaminants, improve their compatibility with HDPE resins and improve current sorting and recycling processes to facilitate success in the above issues.	Alzerreca et al., 2015
Unknown, Iran	HDPE and PP	Wood-plastic composites	Mechanical characterization through flexural, tensile and impact testing was performed in varying contents' specimens of both virgin and recycled HDPE and PP mixtures with sawdust.	PP (virgin and recycled) composites were found to be stronger and stiffer than the HDPE ones (virgin and recycled), although PP has increased mechanical properties in its virgin form. Substitution of virgin polymers by their recycled equivalents was found to be feasible as mechanical properties were comparable in virgin and recycled plastics. Composites made of mixed plastic (both recycled HDPE and PP) were found to have superior properties.	Najafi et al., 2005

Unknown, United Kingdom	HDPE	Unknown	Mechanical characterization through tensile and impact loading was performed and changes in the stress–strain characteristics, yield stress, tensile strength, and tensile (Young's) modulus, of ductility and toughness, all as a function of fiber content were determined experimentally in a composite laminate of a natural flax fiber and recycled HDPE and compared to the ones of a flax fiber-virgin PP one.	Although no comparison was made between a similar composite made of virgin HDPE, properties in the recycled HDPE composite were found to have similar mechanical properties as the ones in the flax fiber-PP one. A volume fraction of between 15 and 20% flax fiber was found to be maximize the mechanical properties measured.	Singleton et al., 2003

Annex E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Projecto n.º P51
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Folha 1 de 1