Plastic packaging sector: a contemporary and crucial challenge

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Thesis submitted for examination for the degree of Master of Science in Technology.

Tokyo 29.7.2020

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Abstract of the master's thesis

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Title Plastic packaging sector: a contemporary and crucial challenge

Degree programme Master of Science (Technology)

Major Advanced Materials for Innovation and Sustainability (AMIS) **Code of major** SCI3083

Supervisor Jaakko Timonen

Advisor Jaakko Timonen

Date 29.7.2020 **Number of pages** 116 **Language** English

Abstract

Recently, the packaging sector started to aim at the reduction of the ecological impact of one of its main materials: plastic. Pictures of plastic marine litter among others democratized bad reputation of this material. Hence, alternatives and bans started to appear. However, plastic displays various important properties for content protection, which are difficult to reach by using alternative materials at industrial scale. In the past, several materials were supposed to replace synthetic plastic in packaging, including polylactic acid, without success. The usages of plastic packaging make recycling complicate, and thermal recovery plants and landfills are still widely used to process these plastic wastes. Nevertheless, innovations have the potential to change this sector. Among materials that could replace plastic, nanocellulose appears as a solid competitor. Regarding recycling technologies, the recent discoveries of plastic enzymatic degradation could solve contamination and diversity problems encountered with plastic packaging wastes.

Keywords plastic, packaging, recycling, materials, technologies, innovations

Preface

My master thesis took place in DIC Corporation, in Japan, during the COVID-19 crisis of 2020. Hence, its progress was disrupted, and my initial missions were changed. With Jaakko Timonen, my thesis supervisor, we modified the aim of this thesis into a more suitable one, regarding this exceptional and unpredictable situation. Thus, the aim of this paper is to present the current state of the plastic packaging sector, from materials properties and recycling to innovations. This topic concerns every potential reader, knowing that everyone buys or uses products daily and so packaging indirectly.

I would like first to thank Nicolas Bétin and Kuniko Torayama for making this experience at DIC possible, spending time on administrative procedures, and supporting me during the emergency state in Japan. I also thank my managers and colleagues Fukuda-san, Kido-san, Gen-san and Uemura-san, for their welcoming and kindness. Special thanks to Kido-san, with whom I was in daily contact during the emergency state. With the team, I investigated different packaging aspects and realized factories visits that are included in this paper. I am grateful to my thesis supervisor Jaakko Timonen, who provided the guidelines of this thesis and answered my interrogations. Finally, special thanks to my classmates for their careful rereading, and all the people who will read this paper.

Tokyo, 29.7.2018

Pierre-Alexis BEY

Contents

Symbols and abbreviations

Symbols

$\overline{M}_N$	number average molecular weight
T_m	melting temperature
T_g	glass transition temperature
E	Young's modulus
UTS	ultimate tensile strength
HV	heat value
MVTR	moisture vapour transmission rate
OTR	oxygen transmission rate

Abbreviations

Polymers

PO	polyolefin
PE	polyethylene
HDPE	high-density polyethylene
LDPE	low-density polyethylene
PS	polystyrene
EPS	expanded polystyrene
HIPS	high impact polystyrene
PVC	polyvinyl chloride
uPVC	unplasticized polyvinyl chloride
pPVC	plasticized polyvinyl chloride
PP	polypropylene
PET	polyethylene terephthalate
PEF	polyethylene furandicarboxylate
EVOH	ethylene vinyl alcohol
PLA	polylactic acid
CNF	cellulose nanofibrils

Concepts

MSW	municipal solid waste
NPO	nonprofit organization
EPR	extended producer responsibility
SU	single-use

1 Introduction

Plastic recently got a negative image regarding its ecological impact. Among plastic materials, plastic packages saw a clear decline of popularity. Indeed, plastic bags and bottles are now associated with marine litter and impactful pictures of dead animals. However, there are technical reasons for remaining plastic use in this industry and wastes leakage in the environment. The aim of this paper is to present these reasons and introduce concepts that could improve the current packaging sector.

For this purpose, this paper will often refer to "packaging" and "package". Thus, it is important to define this concept first, in order to avoid any misunderstanding. The Oxford dictionary defines packaging as: "materials used to wrap or protect goods". By using the same dictionary, the proposed definition for package is slightly different. However, they have close meanings and will be considered alike in this report. Hence, packaging and package refer both to a container, without distinction of content. In this paper, packaging and package will equally refer to liquid bottles, bags, films or trays, to name but a few. An important point highlighted by this definition is the notion of protection. This characteristic is central in the packaging material choice. In fact, it would even have been more relevant to define packaging as “materials used to wrap and protect goods”. Protection is not a characteristic among others, but the main one of packaging.

Indeed, plastic packages were created to this end. Packaging can be made with different materials, including glass, paper/cardboard, metals and plastic. Plastic generally refers to a synthetic material composed of polymers. However, it is important to notice that plastic can be bio-based, and that it was the case for the first plastic packaging, cellophane (1908), composed of cellulose [1]. At this time, cellophane became successful by increasing storage time, while allowing a better products exhibition. Later, Coca-Cola company changed its glass bottles for plastic ones, in order to avoid breakage and reduce amount of material used. Plastic in packaging became soon popular and production increased, without enough end-of-use treatment infrastructures and efficient recycling technologies.

Hence, in 2015, worldwide production of plastic packaging wastes exceeded 140 Mt [2]. This production is supposed to increase year after year, following plastic packaging production. In 2017, worldwide plastic production nearly reached 440 Mt, including nearly 160 Mt of plastic packaging [3]. The annual plastic production is supposed to be multiplied by four in 2050. This increase will aggravate plastic littering pollution, particularly linked to plastic packaging sector. At the present time, around 8 Mt of plastic wastes leak into the ocean each year [4]. Plastic packaging wastes represent a huge part of this amount, more than 50% [5]. Indeed, plastic packaging short lifetime, small dimensions and light weight make them more easily escaping the normal wastes path. This leakage is problematic because plastic degrades slowly in natural environment. Indeed, in the ocean, degradation rate of synthetic plastics reaches at most 40 micrometers per year, whereas this rate is nearly nil for several synthetic plastic types [6].

In order to tackle this problem, organisms specialized in packaging sector, including the Ellen MacArthur foundation (cf. 4.1.1), are focusing on the development of the

three R policy. The tree R policy is based on three notions regarding packaging: reduce, reuse and recycle. These three notions are linked. For instance, by reusing and recycling plastic materials from packaging, the amount of necessary raw materials will be reduced. Moreover, by reducing the production of problematic wastes, including single-use packaging, contamination of recycling streams will be reduced, and recycled materials quality improved. Worldwide, many companies already committed to respect this policy (cf. 4.2.1) [7].
Redesign is also often added to the three precedent concepts. Redesign of packaging consists in changing their composition (materials used), but also their shape. For instance, making a package with only one plastic material, and/or avoiding small pieces in packaging design to avoid leakage. However, plastic is not easily replaceable. Indeed, plastics own good barrier and mechanical properties, which are useful for content conservation. This aspect will be presented in the following section.

2 Packaging plastics polymers

The plastic packaging sector is currently constituted of a large panel of plastic types. However, all these packaging plastics belong to the thermoplastics family. Thermoplastics have to be differentiated from thermoset plastics. The main difference is that thermoplastics, when submitted to heat, can be melted again and used to produce new thermoplastics [8]. Thus, they can be recycled by heat treatment. On the other hand, thermoset plastics cannot go through this melting due to their special structure. Hence, they have better mechanical properties, but are hard to recycle through thermal treatment compared to thermoplastics. Thermoplastics diversity in packaging is a huge advantage, but also one of its bigger disadvantage. Indeed, each plastic type has its own properties, more relevant for certain products than for others, and its own recycling conditions. If all plastic packages and products in the world were manufactured with the same type of plastic, the recycling rates would be drastically higher than the current ones (considering that this unique plastic type is chosen among not so difficult to recycle plastics).

The following parts present the five main types of plastics used in the packaging sector. For each type, formation process, properties, main applications and recyclability are described.

2.1 Polyethylene (PE)

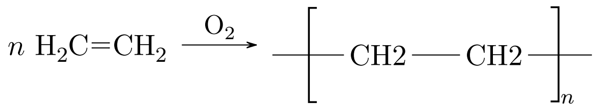
2.1.1 Structure and reaction process

PE is one of the most used polymer in the world. Its annual production exceeds one third of the global plastics market. Moreover, its formula is one of the simplest among polymers. In this part, two subfamilies of PE will also be described: high-density polyethylene (HDPE) and low-density polyethylene (LDPE). These two types of PE represent around 80% of the total PE plastic production. In Western Europe, in 1999, LDPE family represented 34% of the plastics packaging processors consumption, and HDPE 22% [9].

For PE production, polymerization is realized with ethylene monomers. Polymerization is the reaction of single entities, monomers, to create a long chain called a polymer. Different processes were developed through the years in order to obtain different types of PE polymers, with long or short chains, high or low density. Before the 1960's, the main process to create PE in the commercial sector was the high-pressure process. This process consists in polymerization of ethylene monomers. High-pressure polymerization of PE is realized with the following conditions [10]:

- High-pressure, between 1000 and 3000 atm (1 atm $\simeq$ 1.013 bar).
- Temperatures between 80 and 300°C.

It requires ethylene monomers C_2H_4 and oxygen as initiator of the reaction.



$$n\ H_2C{=}CH_2 \xrightarrow{O_2} \left[CH2 - CH2 \right]_n$$

Polyethylene polymerization

One of the main points of this high-pressure reaction is that, at high temperatures, other parasite reactions can occur. These reactions can engender formation of compounds, including hydrogen or methane, and are exothermic. The control and initial conditions of the polymerization are really important in order to keep the reaction under control. The advantage of this reaction is its flexibility. By changing many parameters, including pressure, composition or temperature, polymer attributes can be changed separately. Thus, molecular weight and branching, which are dependant of these parameters, can be modified.

With this process, only PE polymers with a limited number average molecular weight ($\overline{M}_N$) can be produced. $\overline{M}_N$ is defined as the total weight of the polymer, divided by the total number of molecules. If a dedicated polymer is constituted of different types of molecules, with the type index being i, that N_i is the number of i type molecules, and that M_i is the i type molecules weight, then $\overline{M}_N$ formula can be defined as:

$$\overline{M}_N = \frac{\sum_{i=1}^{\infty} N_i M_i}{\sum_{i=1}^{\infty} N_i}$$

In fact, there is a direct relation between $\overline{M}_N$ and the density of a polymer. The higher the number average molecular weight, the higher the density, and vice versa. The range of $\overline{M}_N$ for PE family is 10,000-40,000 $g.mol^{-1}$ [10]. Polymers created with this high-pressure process are called low-density polyethylene (LDPE). The density is in between 0.91 and 0.94 g.cm^{-3}.

At the end of the 1950's, the Ziegler process was developed for the synthesis of polyethylene. This process is also known as co-ordination type polymerisation. Indeed, it requires the use of a catalyst complex in order to orientate and control the monomer attachment to the main chain of the polymer [11]. In general, the catalysts used in the industry for the Ziegler process, as catalyst complex, are the tetrachlorotitanium ($TiCl_4$) and triethylaluminium ($Al(C_2H_5)_3$). This catalyst complex can be prepared before or the two elements can be directly introduced in the main reactor. The conditions of the Ziegler process are [10]:

- Temperatures around 70°C (always under 100°C).
- No water or oxygen to keep the catalyst effective.

Thanks to this process, PE polymers whose density is around 0.95g.cm^{-3} can be obtained. Those PE polymers are the high range of high-pressure process polymers and can be considered as HDPE.

In order to obtain HDPE with density higher than 0.95g.cm^{-3}, Phillips process can be used. In this process, ehtylene monomers are dissolved in cyclohexane or another liquid hydrocarbon. A metal oxide catalyser is used. The reaction conditions are [10]:

- Temperatures in between 130 and 160°C.
- Chamber pressure in between 14 and 35 bar.

The Phillips process allows to recover PE polymers with a density around 0.96g.cm^{-3}, the highest density for commercially produced PE.

2.1.2 Properties

Because polyethylene is a wide family, with many different sub-types of polymers, this part will focus on the properties of low-density and high-density polyethylenes, the two main PE polymers used in the plastic packaging sector. LDPE structure differs from HDPE one [12]. On one hand, low-density polyethylenes present a chain structure with side-branches, creating a disorganized structure. On the other hand, high-density polyethyelenes chains are regularly organized, without side-branches (Figure 1). This difference of structure is the reason of these different densities. Because LDPE structure is disorganized, the chains take more place, and the density is lower, whereas organised chains of HDPE use less space, and the density is higher.

Figure 1: Structures of HDPE and LDPE

The more a polymer has an organized structure, the more it can be described as *crystalline*. The more it has a disorganized structure, the more it can be described as *amorphous*. Because of that, HDPE is more crystalline than LDPE, and there are stronger interactions between HDPE chains than LDPE ones. crystallinity of low-density PE is in the range 40-65% and around 90% for high-density PE. This difference of structure has an impact on the properties of these polymers. However, both LDPE and HDPE belong to the semi-crystalline polymers family.

All the polymers which have crystalline behavior are not totally crystalline (Figure 2), they also own amorphous areas. Thus, the name "crystalline polymer" refers in fact to a semi-crystalline polymer. But this semi-crystalline behavior benefits manufacture of plastics. Indeed, crystallinity gives a strong behavior to polymers, but also a brittle one [13]. The presence of amorphous areas, even if limited, allows polymers to be strong, but also to have toughness. They can absorb impacts and deform. Another phenomenon related to crystallinity is the rotation of molecules. Rotation of molecules takes place around single bonds, including C−C bonds in PE.

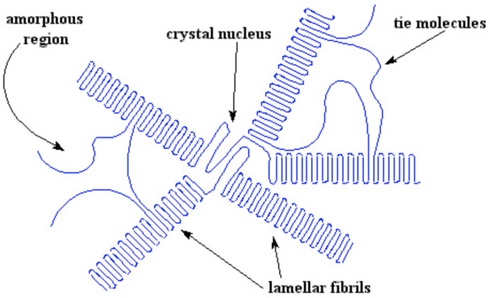


Figure 2: Semi-crystalline state

Above a certain temperature, defined as the glass transition temperature (T_g), these rotations of molecules become limited. Above this temperature, there is a flexibility among chains, which are able to slide and move if a sufficient force is applied. The polymer moves from a glassy and rigid state to a soft one. This state is reversible, and if the temperature decreases below T_g, the polymer state comes back to a rigid state, with molecular rotations. All polymers, semi-crystalline or amorphous, have a glass transition temperature (Figure 3).
This is not the case of the melting temperature (T_m), from which the chains of polymers start to deteriorate irreversibly. T_m defines the temperature from which the crystalline order of a polymer is deteriorate. Because of that, only semi-crystalline polymers have a melting temperature. By definition T_m is always higher than T_g.

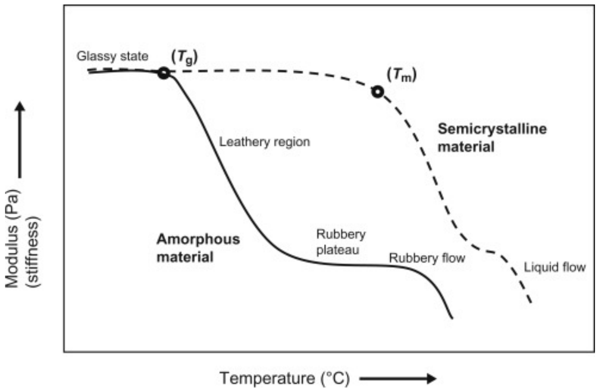


Figure 3: Polymers behavior in function of temperature [14]

The glass transition temperature of PE is under debate for many years. This temperature is placed in between -130 and +60°C according to references. However, the more recurrent value is T_g = -120°C. Concerning melting temperature, they are slightly different according to the density of PE polymer [15]:

- LDPE: T_m = 110°C.
- HDPE: T_m = 130°C.

The difference in chains organization between LDPE and HDPE has also an impact on the mechanical properties of these polymers. Because HDPE has a higher crystallinity,

it has higher hardness and a lower viscosity compared to LDPE. Low-density PE will have a higher impact strength than high-density one.
There are numerous mechanical and barrier properties concerning polymers, more or less relevant concerning plastic packaging sector. This paper will focus on four mechanical and barrier properties of polymers: *Young's modulus*, *tensile strength* (and so *elongation*), *impact strength* and *oxygen and water transmission rates.*
Young's modulus is used to describe and quantify the stiffness of a material, which is the resistance to deformation under an applied force. It can be described with Hooke's law formula as,

$$E = \frac{\sigma}{\epsilon}$$

with E being the Young's modulus, ϵ the deformation along a precise axis and σ the stress applied along the same axis. ϵ is itself defines as the ratio in between the length deformation (ΔL) and the length at rest (L_0). The Young's modulus can then be redefined as:

$$E = \frac{\sigma . L_0}{\Delta L}$$

The Young's modulus depends on the density and crystallinity of the polymer. Indeed, it is not the same for the different types of polyethylene.

- LDPE: E varies from 0.2 to 0.3 GPa.
- HDPE: E ranges from 0.6 to 1.5 GPa [15].

This difference between the two PE types is limited but understandable. The amorphous zones are present in a higher proportion in LDPE. As depicted in Figure 2, these zones are the ones in which chains density is lower, where chains are less ordered. When applying a stress, deformation will require chains moving. If there is more amorphous space in the polymer, it is easier to deform it as a lower stress is needed.

Tensile strength is defined as the capability to resist a pulling stress. But commonly, the term "tensile strength" is used to refer to ultimate tensile strength (UTS), or breaking strength, which is the maximum pulling stress that can be applied to the material before it breaks. This data is interesting, because depending on the structure of the material, the behavior of the tensile strength curve will be drastically different. Amorphous materials will have a lower UTS than brittle ones, but they will be able to deform more before breaking. Figure 4 shows the tensile strength evolution in between HDPE and LDPE. The crystallinity difference has a great impact on the behavior of these polymers under a progressive applied stress. In this experiment, HDPE density was around 0.95 g.cm^{-3} and LDPE one 0.92 g.cm^{-3} [16].
The ultimate tensile strength values according to this experience are:

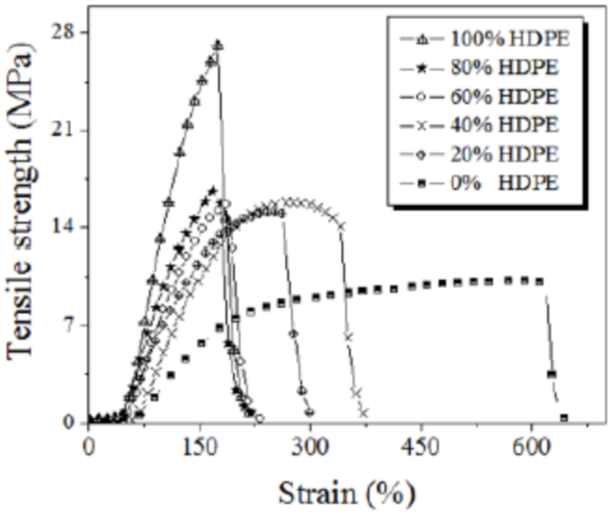


Figure 4: UTS difference between HDPE and LDPE using mixtures [16]

- LDPE: 9.9 MPa.
- HDPE: 27.9 MPa.

HDPE, with a higher crystallinity, has a limited deformation range. The relation between stress and strain is linear until break of the material. It depicts the brittle behavior of the polymer, with a limited deformation (up to 150%). On the other hand, LDPE and its structure more amorphous allows a higher deformation (up to 600%), but only for moderate applied stress. For ductile materials, this linear area, called elastic behavior, is limited and followed by a plateau, or sometimes a decrease of the stress for an increasing strain. The last point of the linear zone is called the yield point, linked to the yield stress. It is the limit between the elastic zone (linear) and the plastic zone. As long as it stays in the elastic zone, the polymer or material will be able to regain its original shape. But when it goes in the plastic zone, stress applied starts to deteriorate the structure, and the deformation is irreversible. For brittle materials, the yield stress and UTS are usually close.

Impact strength is the capacity for a material to resist an abrupt load. If the applied load norm is higher than the limit one, it will break the material. This information is particularly relevant in the case of packages, that can be subject to hard conditions during transport or storage. The impact strength is measured originally in foot-pound force, according to the test used to measure it, but is here converted in $J.cm^{-1}$:

- LDPE: 13.5 $J.cm^{-1}$.
- HDPE: 4.3 $J.cm^{-1}$ [15].

These values are coherent with the structure and other mechanical properties of these polymers. Indeed, as depicted before, due to its higher crystallinity, HDPE is not able to deform as much as LDPE. This capacity to deform is important to absorb the energy transmitted during the impact. Because of the lack of amorphous spaces in the HDPE structure, this absorption by deformation is weaker than for LDPE.

Oxygen transmission rate (OTR) quantifies the capability of a material to diffuse oxygen, its permeability to oxygen molecules. It refers to the amount of gas that go through for a given period. This parameter will depend on the structure of the material, and the way it complicates the path of gas molecules through the material. Values for both PE studied here are taken at 30°C. To quantify this value, the *barrer* unit can be used. This unit is not part of the SI system, and 1 barrer equals to $10^{-10}.cm3(STP).cm/(cm^2.s.cmHg)$.

- LDPE: 5.5 barrer.
- HDPE: 1.1 barrer [10].

The OTR of LDPE is higher than for HDPE, which means that for the same experiment conditions, a higher amount of oxygen molecules will cross the low-density polymer. The structure has again a huge impact on this property. Indeed, amorphous zones of PE polymers act as free volumes, where the oxygen molecules path is easier [17]. LPDE has a higher proportion of amorphous zones than HDPE, thus the OTR of the low-density PE is higher. Typically, the ratio of high-density over low-density PE OTR is around one fifth.

The same principle of transmission rate can be defined with gaseous water. It is often called water vapour transmission rate (WVTR) or moisture vapour transmission rate (MVTR). It is the transmission rate for a given period of the water vapour through a material. The unit for this measure is $g.m^{-2}$.

- LDPE: around 19 $g.m^{-2}$.
- HDPE: around 5 $g.m^{-2}$ [18].

As for the OTR, the difference in between LDPE and HDPE is due to the proportion of crystalline zones, that do not allow water molecules to go through (or with more difficulties).

	E (MPa)	UTS (MPa)	Elongation (%)	Impact strength ($J.cm^{-1}$)	OTR (barrer)	WVTR ($g.m^{-2}$)
HDPE	1,500	27.9	150	4.3	1.1	5
LDPE	300	9.9	600	13.5	5.5	19

Table 1: HDPE and LDPE properties

2.1.3 Main applications

As predicted, main applications of a polymer will depend on its properties. The cost is also an important factor, but it will not be discussed in this report. The differences in PE types properties, highlighted in the previous part and Table 1, are sufficient to differentiate the uses and applications of LDPE and HDPE. The first one appears to be more flexible and deformable, with a higher impact resistance and better impermeability for both water vapour and oxygen. The second one is more brittle, but it is able to retain its shape under higher applied stress. The main applications of these PE types among the packaging sector are then split as follows [19]:

- LDPE: films and wraps (shrink wraps); squeeze bottles; and shop and rubbish bags.
- HDPE: bottles caps; milk, juice and chemical bottles (among others); food containers (the most known being ice cream container).

2.1.4 Products recyclability

According to the main applications of the polymer material (presented above), it could be more or less difficult to recycle it. Products require sometimes coatings or additives that can interfere with the normal recycling process of the material. This part shows both LDPE and HDPE products requirements for optimal material recycling [20].

- Color of HDPE as body product (bottles or containers): for optimal recycling, HDPE needs to have its natural colour, but light tints colored and opaque products may be suitable for recycling, with a lower recycling rate. Black HDPE is not recycled (cf. 5.1.5).
- Coating on HDPE as body product (bottles or containers): the highest recycling rate is achieved for high-density PE without coating. Ethylene vinyl alcohol (EVOH) is tolerate but only at low proportion. However, recent tests allowed to improve quality of recycled HDPE with high content of EVOH as barrier layer. HDPE products with polyvinylidene dichloride (PVDC) coating cannot be recycled.
- HDPE as closure (caps): good recycling if mono material.

The main contaminant of HDPE material recycling stream is polypropylene (PP). Indeed, this polymer is often associated to HDPE in bottles, as material for cap or body. A classic bottle association is HDPE body and PP cap. Only a small proportion of recycling plants can separate PP and HDPE, thanks to their different melting temperature. For recycled content production, a limit of 5% PP contamination is tolerate, which can reach 10% for recycled content that will be used for coarse applications.

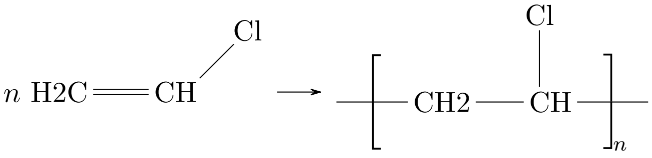
Because LDPE is used mainly in wraps or films, its recycling is challenging due to food contamination and lightweight. But when light-density PE is adopted for other non-contaminant or heavier applications, its recycling process and recycling conditions are similar to HDPE ones.

In the next parts, other types of plastics will be presented. For each of them, same properties and concepts than the ones described in this part will be used. Please refer to the above content.

2.2 Polyvinyl chloride (PVC)

2.2.1 Structure and reaction process

The use of PVC in plastic products has largely increased due to discoveries of additives and stabilizers which helped in the production process. During the 20th century, polyvinyl chloride was, with polyethylene, the main polymer for plastic materials production. But during the 70's, studies found a link in between PVC production plant and several bad diseases, leading to death of many factories employees. After these terrible events, suffering from bad reputations and new regulations, the PVC production slowed down. In 1999, in Europe, this polymer represented only 5% of the virgin plastics used in the packaging sector (around 700 kilotons) [9], far away from the 56% and 7,500 kilotons of HDPE and LDPE combined this year.
PVC is obtained by polymerization of vinyl chloride monomers. Three polymerization processes are used nowadays for the production of polyvinyl chloride: suspension, emulsion and bulk [21]. Polymerization by suspension is the more widely used among the three techniques. More than 80% of PVC is produced thanks to suspension polymerization. It is a free radical polymerization, which means that free-radical monomers are added successively in the mixture in order to form the final polymer. This mixture is placed in the reactor with a liquid phase, often water, and blended to accelerate the polymerization.



Polyvinyl chloride polymerization

For this reaction, temperatures in the range 50-75°C are chosen, which can influence the molecular weight of the final polymer.

Structure of PVC and PE are closed, the difference being that, for polyvinyl chloride, one of the hydrogen atoms is replaced by a chlorine atom. Thus, a dipole is created on the C−Cl bond, which gives a higher polar behavior. Compared with polyethylene and C−H bonds, the electronegativity difference is higher in between C and Cl atoms than in between C and H atoms. The absolute value of this difference is 0.35 for C and H, and 0.61 for C and Cl.

The number average molecular weight $\overline{M_n}$ of commercial PVC polymers is in the range 45,000-64,000 $g.mol^{-1}$. Its density is comprised in the range 1.1 to 1.45 $g.cm^{-3}$ depending on the quantity of additives in the polymer compound (see below) [10].

2.2.2 Properties

By X-ray analyzes, it was shown that commercial PVC is mainly amorphous, with only a small percentage of crystalline structure, around 5% according to these X-ray studies [10]. There is a structural repetition in the organization of these crystalline zones, which are placed 5 Åeach other. The structure of PVC seems to be closed to the one of LDPE presented in the previous section. The exact distribution of branches is still under debate, but recent studies lead toward one long branch per polymer molecule and less than ten short ones, similar to low-density PE branches mechanism.
Because of its amorphous structure, polyvinyl chloride does not have a melting temperature, only a glass transition temperature (Figure 3). This T_g is estimated around 78°C for pure PVC [22]. But as for other properties, it is quite useless to give the glass transition temperature of pure polyvinyl chloride. It could be useful when studying PVC resin itself, but it is useless when looking for packaging application. Indeed, pure PVC, in its massive form, is a basic polymer with limited properties. It is colorless, has a narrow heat stability and, when heated, develops good adherence with surfaces of metallic materials. In order to improve these properties, other compounds need to be introduced in the fabrication process. These compounds will have an influence on the properties of the PVC material. Two categories of polyvinyl chloride can be distinguished: rigid and soft. The main ingredient of rigid PVC is pure PVC resin, more than 75%, whereas for its soft version, this resin accounts for about 50% of the total composition (Figure 5).

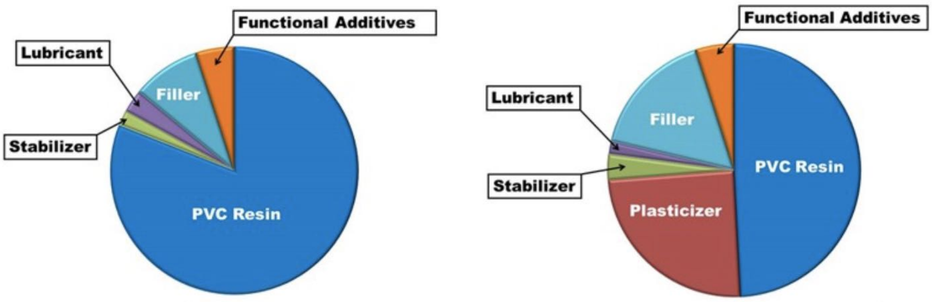


Figure 5: Rigid PVC (left) and soft PVC (right) compositions [23]

The different types of compounds added to PVC resin are:

- Plasticizers, to make polyvinyl chloride compound soft and flexible.
- Stabilisers, to allow PVC compound to support tougher environment. Main categories are heat and UV stabilisers.

- Fillers, to enhance properties of the polymer, including impact and tensile strengths.

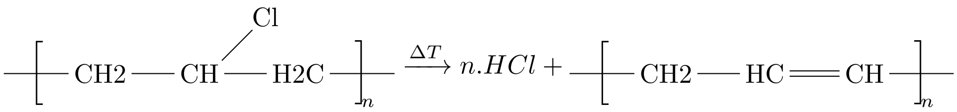
These are the main types of additives, but others exist that can be used depending on the wanted final product, as lubricants, which are introduced to reduce adhesion problems during manufacturing, or extenders, used to replace part of plasticizers in the process and reduce total costs. Indeed, extenders are less expensive than true plasticizers. But this part will highlight only the following three types: plasticizers, stabilisers and fillers, because of their direct impact on the product properties.

The annual consumption and production of plasticizer materials is huge and exceeds the one of numerous polymers. More than 1,400 types of plasticizers are registered in the dedicated database [24]. This impressive number of plasticizers allows the existence of a dedicated additive for nearly each requirement. But these products have also adverse effects, among them color and toxicity impacts. They need to be chosen carefully, according to the final application. Usually, more than one plasticizer is used to enhance and improve PVC properties. Among all these products, the main category used is phthalates, especially diisodecyl and dioctyl phthalates. They are the first choice of PVC producers because of their good efficiency and low cost. After years of use, they are now considered as default choice and researches on greener alternatives slowed down.

As described before, PVC is a polar molecule due to the C−Cl bond. Plasticizers use polarity and good solubility to be linked and take place inside the polyvinyl chloride structure [25]. Plasticizers take place inside the amorphous regions of the polymer, creating space in between polymer chains. This space engenders a decrease of the glass transition temperature of PVC compounds. Indeed, cohesion and interaction in between polymer chains are weaker, and the thermal stability is affected. The main reason to introduce plasticizers in PVC compounds is to make it more flexible and soft, in order to diversify its applications. Thus, the impact on mechanical properties depends on the nature of the plasticizer used and its percentage in the total composition. But usually, an increase of plasticizers content engenders an increase of the elongation at break and a decrease of the UTS.

Stabilizers are mainly used to increase the heat stability of the polymer. It is an essential compound knowing the poor thermal stability of PVC. Indeed, when heated at a temperature range higher than 70°C, numerous adverse effects can appear. However, processing temperatures are often around 200 °C, which can creates problems with PVC. Degradation will occur first on the color of PVC, at the beginning translucent white, which will turn yellow and even black. Mechanical properties will also be affected after color deformation, if the degradation is too important. But for a commercial use, this color criterion is as important as the bulk properties. For these reasons, stabilizers are used in order to limit degradation during fabrication process and obtain a correct and usable product.

When heating this polymer without stabilizers above its glass transition temperature, thermal degradation phenomenon always occurs. When the temperature exceeds around 70°C, the first loss of HCl appears in the polymer structure:



$$\left[-\mathrm{CH2}-\overset{\mathrm{Cl}}{\mathrm{CH}}-\mathrm{H2C}- \right]_n \xrightarrow{\Delta T} n.HCl + \left[-\mathrm{CH2}-\mathrm{HC}=\mathrm{CH}- \right]_n$$

But this first loss engenders loss of neighbors Cl, and formation of more HCl. This chain reaction is called zip dehydrochlorination [26]. This degradation propagates along the back bone of the polymer. Reasons for this thermal degradation process are still under debate in the scientific community. In terms of stabilization effects, the more efficient and used thermal stabilizers for PVC are lead salts, but they could not be the best option considering environmental and toxicity impacts. For this reason, researches in the field of "green stabilizers" are conducted. The main property of a PVC thermal stabilizer should be to nullify and swallow HCl, responsible of thermal degradation [27]. For that, primary and secondary stabilizers can be introduced. The role of primary stabilizers is to react with allylic chlorine atoms. These atoms are created just before the formation of HCl molecules. These primary stabilizers must own a high reactivity. But this high reactivity is double-edged. If formation of HCl happens, these primary stabilizers will react with HCl instead of allylic chlorine atoms. To avoid this problem, secondary stabilizers are introduced, whose role is to react with HCl. Thanks to these secondary stabilizers, primary ones can operate without disturbance by unwanted reaction with HCl.

In order to keep a commercial and use interest, these additives need to have an impact as low as possible on the other properties and general aspect of the polymer, for instance not alter PVC color. The cost is also important in the choice of the final additive.

Other additives used for PVC compounds are fillers. Fillers are the additives used to reduce cost and enhance diverse properties of the polymer. They take place in the polymer matrix, filling spaces in between polymer chains. They can modify electrical to mechanical properties.

PVC polymer is known for its thermal degradation, mentioned in the above paragraph, but also its low impact strength. This property can be improved with calcium carbonate ($CaCO_3$) as filler. This additive will be introduced in the polymer matrix and will increase the impact strength capacity. But particles size needs to be chosen carefully. It was proven that a too large $CaCO_3$ particles size could have the inverse effect and decrease impact strength of PVC. The reason is a poorer adhesion between calcium carbonate particles and polymer matrix. An adequate $CaCO_3$ particle size (under micron) provides an enhancement of impact strength for polyvinyl chloride compounds [28].

There are numerous fillers with different effects on polymer properties. For instance, talc can be used to increase stiffness, and metal and carbon fibers have an impact on abrasion resistance. However, it is necessary to choose adequate fillers and to know all their effects on the polymer properties, in order to avoid unwanted effects or contradictory ones. For instance, Mica is known as permeability enhancer, but it has also an impact on elongation at break.

PVC properties depend on the additives present in the polymer compound. Thus, it is complicate to depict precisely the value or even the values range of the basic properties. However, common values for commercial grades of both plasticized and unplasticized PVC will be given, in order to compare the commercial applications with other plastic materials highlighted in this section. Rigid PVC has no plasticizers, it is called unplasticized PVC (uPVC or PVC-U), and flexible PVC, containing plasticizers, is called plasticized PVC (pPVC or PVC-P). Both are considered as amorphous polymers, thus they do not have melting temperature.

- uPVC: T_g is about 80°C.
- pPVC: T_g is about 50°C [10].

Plasticizers are responsible of this decrease in the glass transition temperature. They are present between the polymer chains, in the amorphous zones, reducing the interaction force of these chains (Figure 6).

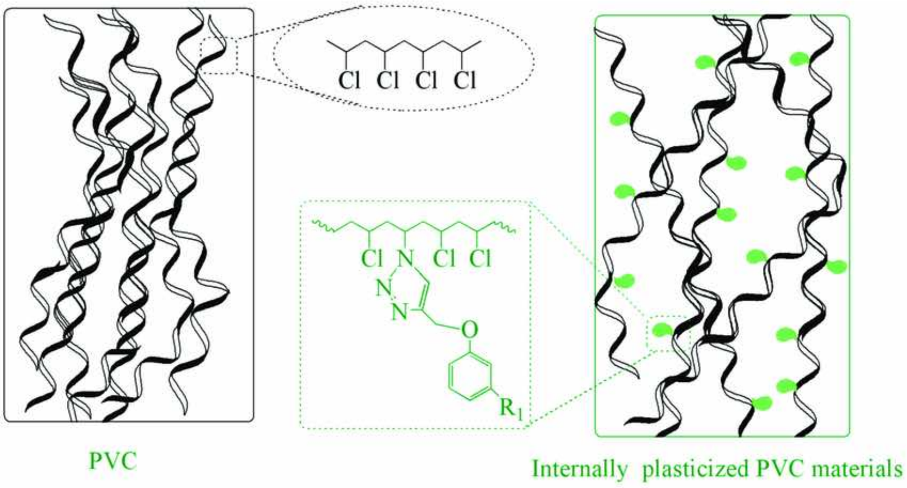


Figure 6: Plasticizers in PVC structure [29]

The same effect happens for other properties, summarized in Table 2. Even if both forms of PVC are considered as amorphous, the crystallinity of pPVC is lower than for uPVC because of plasticizers. Sometimes, uPVC can be considered as slightly crystalline.

	E (MPa)	UTS (MPa)	Elongation (%)	Impact strength ($J.cm^{-1}$)	OTR (barrer)	WVTR ($g.m^{-2}$)
uPVC	3,000	50	80	0.6	0.12	15 to 80
pPVC	100	20	400	6	-	

Table 2: PVC properties [10][18][30]

When used as food film/wrap, compared to other common polymers, PVC has a lower oxygen permeability [31].

2.2.3 Main applications

After revelations concerning toxicity of PVC process for workers during the 20^{th} century, its production could have been stopped. But as mentioned in the precedent part, polyvinyl chloride compounds have an advantage compared to other commercial polymers: diversity among properties. This diversity is due to the higher number of additives that can be used with PVC, mainly because of its polar nature. Logically, this diversity can be found in the diverse applications of PVC, from construction with pipes (first market for this polymer) to health sector, with packaging sector being the second most important market, with 3 Mt per year [32].
In the past, regarding packaging applications, PVC was the leader in the sector of food trays and beverage bottles. However, it was then replaced because of the too strong concurrency of polyethylene terephthalate (PET), which has a better CO_2 permeability. But PVC is still used for oil bottles, where carbon dioxide permeability is not among the most important criteria. The main applications of PVC depends on the polymer nature. If polyvinyl chloride is more rigid, its applications will be different than for flexible forms. This rigid behavior, as explained above, is due to the presence and amount of plasticizers in the PVC compound.

- uPVC: rigid films (accounting for 60% of total production of PVC for packaging applications), bottles, containers and trays.
- pPVC: cling films (accounting for 11% of total production of PVC for packaging applications) and cap seals for food and beverage packages [19].

Nowadays, the main application for vinyl chloride family is films. It can be used for the protection of fresh food from oxygen and moisture degradation.

2.2.4 Products recyclability

As for other plastics, recycling of PVC is a recent process. For many years, this plastic has been put in landfills. But bans in European countries and the United States spelled the end of this usage and the beginning of material and thermal recycling, especially in these precursor countries.
It is interesting to notice that polyvinyl chloride packaging applications are limited (in 1999, fifth in terms of virgin plastic production in Europe for packaging, representing only 5%), whereas this polymer production is the second most important in the world, just after PE. In countries with adequate recycling plants and infrastructures, PVC can be recycled. However, this recycling process has to be controlled. The nature of polymer engenders impossibility of several methods of recycling, including thermal recovery and compost. This is due to the chemical formula of polyvinyl chloride. As described before, thermal degradation of PVC is associated with formation of HCl. This compound is dangerous, and because the quantity produced is not negligible, thermal recovery is not recommended for PVC recycling [33]. Oxidative degradation of this polymer in the ground, with compost or landfill, is under debate. Indeed, knowledge about risky elements produced during this reaction is too low,

and consequences on the environment could be problematical. For the same reason, most of recycling associations consider this material as not recyclable for packaging applications [34]. Its presence could contaminate the stream of recycling.

Even if recycling of PVC is possible, it is rare for post-consumer wastes. Though, PVC post-consumer wastes represent more than 90% of PVC wastes. As explained before, this polymer is the second most produced in the world, but only a low portion is turned into packaging applications, which will engender post-consumer wastes. Most of the PVC applications are long-term ones, including pipes and window frames. When these products become wastes, they enter in a dedicated waste stream, favoring the recycling of PVC, to create new long-term products. For packages made with PVC, recycling is still complicate, because of the likely contamination of other polymers due to HCl formation. Moreover, PVC compounds contain many additives, sometimes hazardous ones, which make difficult to obtain a clear and safe recycled product.

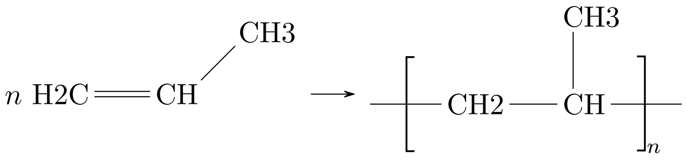
Improvements in the field of PVC recycling are critical. In the next few years, long-life products will become wastes, and it represents more than half of the PVC products.

2.3 Polypropylene (PP)

2.3.1 Structure and reaction process

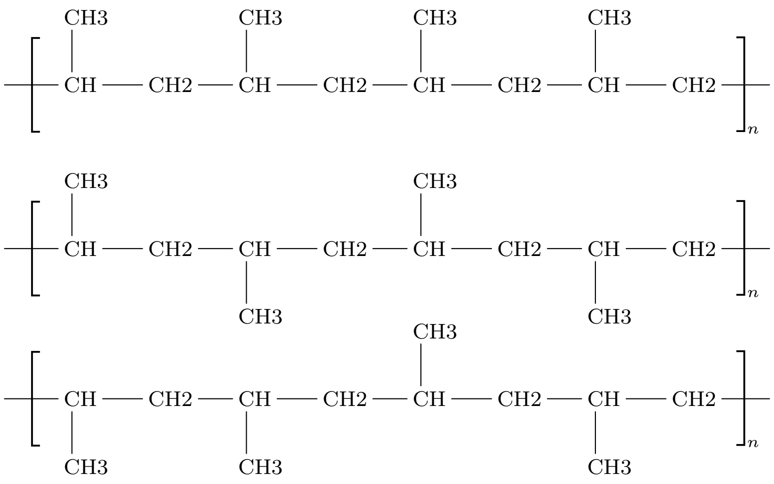
Polypropylene family contains different types of polymers. This diversity is possible by playing on the catalysts used in the reaction process. Every type has different properties. For instance, isotactic PP is similar to HDPE, with changes for few mechanical and thermal properties, whereas atactic PP is amorphous. Polypropylene is the third more produced polymer in the world, after PE and PVC. In Europe, in 1999, it represented around 20% of the production of plastic for packaging sector, second behind PE [9]. However, in several countries in the world, PP represents the first plastic resin used for the packaging sector. For instance, this is the case in the US, with an annual production of 2.3 Mt in 2010.

The formation of polypropylene polymer is realized by polymerization of propene monomers. The production is similar to PE one by using Ziegler catalysts. Naphta is used as diluent of the reaction. It is introduced in the tank with propene monomers and catalysts compounds. The reaction is conducted at a temperature of 60°C, for a duration in the range 1 to 4h [10]. As for PE production, by varying the experimental conditions, including temperature, catalyst types or concentration, molar weight of the final product can be modified.



Polypropylene polymerization

During the polymerization, three forms of polypropylene can be created depending on the catalysts used for the reaction. These forms are isotactic, syndiotactic and atactic ones [35]. The difference between these three types is the organization of the methyl ($-\mathrm{CH}_3$) blocks along the back-bone chain of the polymer (see below). In the case of isotactic polymer, all the $-\mathrm{CH}_3$ groups lie on the same side. For syndiotactic one, there is an alternate "one out of two" layout, whereas for atactic PP, the layout is random. This difference in the layout has an impact on the thermal and mechanical properties. In fact, it has an impact on the general crystallinity of the polymer. When a polymer has a tactic structure, it is more ordered. It allows a better chains arrangement, and reduces the amorphous zones. In the case of polypropylene, both syndiotactic and isotactic forms will have a higher crystallinity than atactic polypropylene, which will be considered as an amorphous polymer. This tacticity behavior is considered as the main difference, due to the methyl group, between PE and PP.



$$-\left[\overset{\mathrm{CH3}}{\overset{|}{\mathrm{CH}}}-\mathrm{CH2}-\overset{\mathrm{CH3}}{\overset{|}{\mathrm{CH}}}-\mathrm{CH2}-\overset{\mathrm{CH3}}{\overset{|}{\mathrm{CH}}}-\mathrm{CH2}-\overset{\mathrm{CH3}}{\overset{|}{\mathrm{CH}}}-\mathrm{CH2}\right]_n-$$

$$-\left[\overset{\mathrm{CH3}}{\overset{|}{\mathrm{CH}}}-\mathrm{CH2}-\underset{\mathrm{CH3}}{\underset{|}{\mathrm{CH}}}-\mathrm{CH2}-\overset{\mathrm{CH3}}{\overset{|}{\mathrm{CH}}}-\mathrm{CH2}-\underset{\mathrm{CH3}}{\underset{|}{\mathrm{CH}}}-\mathrm{CH2}\right]_n-$$

$$-\left[\underset{\mathrm{CH3}}{\underset{|}{\mathrm{CH}}}-\mathrm{CH2}-\underset{\mathrm{CH3}}{\underset{|}{\mathrm{CH}}}-\mathrm{CH2}-\overset{\mathrm{CH3}}{\overset{|}{\mathrm{CH}}}-\mathrm{CH2}-\underset{\mathrm{CH3}}{\underset{|}{\mathrm{CH}}}-\mathrm{CH2}\right]_n-$$

Isotactic (top), syndiotactic (middle) and atactic (bottom) forms of polypropylene

The most ordinary configuration for polypropylene is the isotactic one, with all the methyl groups lying on the same side of the back-bone chain. Because this form is the most used in the packaging sector, this paper will focus on properties of this configuration.

The number average molecular weight $\overline{M_n}$ of commercial polypropylene polymers is ranging between 40,000 and 60,000 $g.mol^{-1}$. It has the lowest density among common polymers used at the same scale, in the range 0.89 to 0.92 $g.cm^{-3}$ [10].

2.3.2 Properties

Perfect isotatic PP exists, but commercial grades of this polymer have a lower crystallinity. The structure of the commercial polymer contains part of atactic PP. The percentage of atactic content in the final product depends on the catalysts and the experimental conditions. The isotactic form represents an important part of the polymer, in a range of 90 to 95%. Depending on this percentage, the general properties of polypropylene will change [36]. The rest of the structure is composed of atactic and syndiotactic PP forms. These forms can be present in the structure as complete polymer chains, filling amorphous space in between isotactic chains, but also as block inside the main isotactic chains. In this last case, a percentage of the total chain is not under the isotactic pattern, but atactic or syndiotactic one.

Both glass transition and melting temperatures are influenced by the isotactic ratio of the polypropylene compound. As explained in the PE part, the crystallinity has an impact on both temperatures (T_g and T_m). Indeed, the higher the crystallinity degree of a polymer, the more the chains are ordered and difficult to move by increasing temperature.

- T_g: -20 to -5°C.
- T_m: 160 to 175°C [36].

This difference in the isotactic form percentage of commercial polypropylene has also an impact on the mechanical properties (Table 3).

E (MPa)	UTS (MPa)	Elongation (%)	Impact strength ($J.cm^{-1}$)	OTR (barrer)	WVTR ($g.m^{-2}$)
690 to 860	28 to 32	around 50	around 0.4	2.3	3.9

Table 3: PP properties [10][18]

The low impact strength of PP is among its more characteristic properties. It is a witness of the high brittleness of this polymer. In order to decrease the brittle behavior of polypropylene, tests have been realized. By using mixture of polypropylene and another polymer, it was possible to get an important increase of the impact strength. When this possibility was discovered, focus was made on PP and ethylene blends, for instance polypropylene-ethylene propylene diene (EPD) rubber, a blend with a drastic increase of impact strength compared to single PP [37].

As for polyvinyl chloride, additives are required to improve properties of polypropylene. However, the proportion of additives used for PP is far lower than for PVC. One important additives for polypropylene is antioxidants. Compared to PE, PP encounters a faster and a more important oxidation at relatively high temperature.

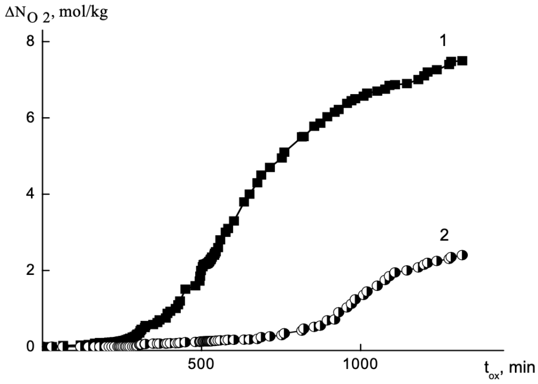


Figure 7: Oxidation of PP (curve 1) and PE (curve 2) at T=110°C [38]

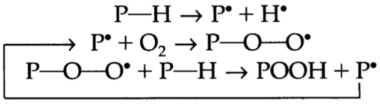


Figure 8: Oxidation reactions of PP [39]

This oxidation (Figure 8) takes place inside the amorphous zones of the polymer structure. The difference between PE and PP degradation (Figure 7) is due to the conformation and the degradation phenomenon. It propagates inside a same molecule for PP, along the chain, whereas it takes place mainly in between molecules for PE.

2.3.3 Main applications

Regarding what was explained above, the main concurrent of PP on the plastic products market is HDPE. They have both a good crystallinity, and their properties are close. Whereas polypropylene has a better heat resistance, with T_g and T_m higher than for high-density polyethylene, and a lower density, which allows to create more plastic products for the same compound mass, its properties are less advantageous for the packaging sector than HDPE. Indeed, it has a lower oxidation resistance and impact strength, two properties important for packaging, in order to protect their content. This drawback can be improved by adding antioxidants to the PP compound, but it increases the cost of this polymer production, which decreases its commercial interest. Recent studies have investigated natural antioxidants to decrease thermal oxidation of PP [40]. These additives are extracted from wastes of tomatoes and grape fruits, known for their natural antioxidant properties. This process could allow to reduce the cost of antioxidant additives for PP while, in the same time, solving an existing waste problem.

Despite these disadvantages, compared to HDPE, PP is considered as an enduring material for several packaging applications. Nowadays, its main applications are for sauce tubs and heat resistant packaging [19]. Concerning this last use, it is one of the reasons why PP is the main material used for microwavable containers. The other reason to use this polymer is that its structure makes it transparent to microwaves, which means that it does not absorb microwaves energy [41]. The association of its heat resistance and transparency to microwaves makes PP a good material for microwavable containers. The content temperature can become high due to microwaves energy absorption, thus the container needs to support these high temperatures without deforming. PP is also used for bottle cap and plug, in association with HDPE.

2.3.4 Products recyclability

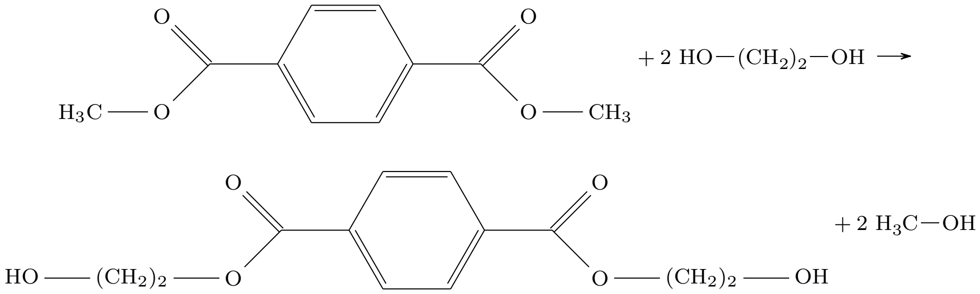
As discussed above in the recyclability part of HDPE, the association of both PP and HDPE in bottles production is frequent. One polymer is used for the body while the other one is used for the cap. Generally, HDPE is used for the bottle body and PP for the top. The reasons for choosing one rather than the other depend on the content, the use and the producer wish. Many parameters need to be taken into account, including the price. However, it is a fact that their mixing creates problems for products recycling. As for PVC, the main challenge with PP recycling is that there is no real standard for packaging made with this polymer. Even when collected separately and free of contamination, issues remain regarding the recycling of PP. Indeed, they are various compositions and colors among PP wastes. Thus, it is complicate to obtain a recycled mixture that is commercially advantageous to use, especially for the color. Moreover, the thermal degradation of PP is a problem during the recycling process, because it affects the properties of the recovered product and so decreases the commercial interest and value. Even if PP represents the main plastic packaging used in several countries, its recycling rate stays really low. In the US, the recycling rate of PP is under 1% [42].

2.4 Polyethylene terephthalate (PET)

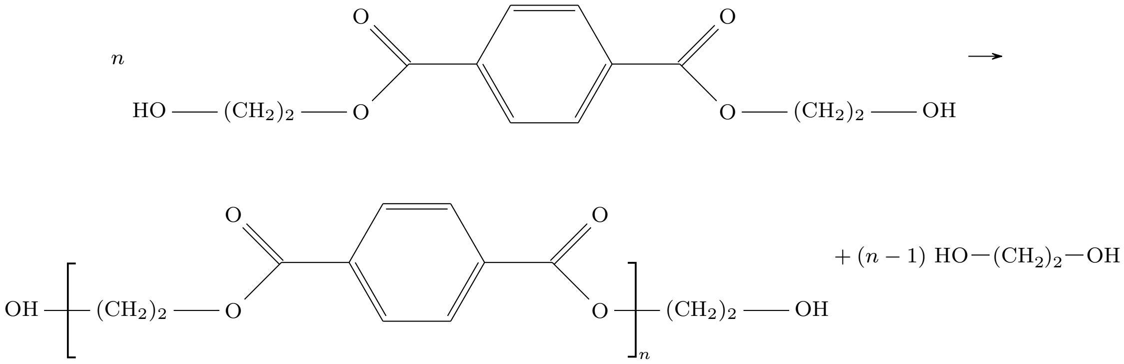
2.4.1 Structure and reaction process

This polymer knew a big increase in its production during the 80's. It was mainly due to the change of material for bottles production, from glass to plastic. For instance, Coca Cola brand introduced PET bottles for the first time in 1978, for its two liters jar, and then decided to extend this material to the whole bottles line. At the end of the 90's, the worldwide production of PET was around 17 Mt per year. In 1999, in Europe, PET production for the packaging sector represented around 1.4 Mt and 11% of this market [9].

Compared to the three precedent polymers presented, PET has a more complex structure. PE and PP belong to the polyolefin (PO) family. This family clusters polymers whose monomers chemical formula is C_nH_{2n}. PVC structure is close to PO one, but due to the presence of the chlorine atom, it does not belong to this category. PET is part of the polyester family and maybe its more famous polymer. As the name *polyester* suggests, the PET structure includes ester group. The reaction is not a single polymerisation as for the precedent polymers, involving only one monomer, but a polyesterification, with two main reactants. These reactants are ethylene glycol ($(CH_2OH)_2$) and dimethyl terephthalate ($C_6H_4(COOCH_3)_2$). Two reactions occur in the PET production, in order to realize the global polyesterification: trans-esterification and polycondensation. Trans-esterification occurs at a temperature in the range 150-220 °C depending on the desired crystallinity, and a pressure of 100 kPa. Both $-CH_3$ groups of the dimethyl terephthalate diester are replaced by $-(CH_2)_2-OH$ groups. Polycondensation is realized at a temperature in the range 270-290 °C, and a pressure of 100 Pa [43]. After these two reactions, usable polymer is obtained, but its applications are limited to the fibres sector. In order to get a PET suitable for bottle production, another polymerisation process is required, the solid state polymerisation. This reaction allows to increase the molecular weight of the polymer, necessary condition to produce solid plastic bottles.



Trans-esterification of ethylene glycol and dimethyl terephthalate



Polycondensation leading to PET polymer formation

These two reactions, trans-esterification and polycondensation, are quite similar regarding the mechanism. However, they required different catalysts. The reason is that catalysts used for the trans-esterification are also catalysts of the decomposition of ester. Moreover, catalysts of the polycondensation are not strong enough to catalyze the trans-esterification. Hence, two separated catalysts are required [44]. For the esterification of dimethyl terephthalate, zinc and manganese can be used among other compounds. For the polycondensation, antimony is used. This last element is known as toxic for the environment and human health. This is one of the defects of this process, the other one being that the management of two catalysts is complicate to handle. Indeed, it is important to be careful in order to avoid contamination, especially contamination of the trans-esterification catalysers during the polycondensation, that could decompose the ester groups.

The number average molecular weight $\overline{M_n}$ of polyethylene terephthalate is in between 20,000 and 30,000 $g.mol^{-1}$ [10]. Indeed, each unit of this polymer has a molecular weight around 200 $g.mol^{-1}$. Then, the number of units in a PET chain varies from 100 for fibres applications, and 150 for bottles production, after the solid state polymerisation. It has a density of 1.39 $g.cm^{-3}$ [45].

2.4.2 Properties

PET has higher glass transition and melting temperatures than the other polymers presented. This property is due to a characteristic of polyesters which is the great cohesion in between polymer molecules. These intermolecules cohesive forces are created because of the polarity of the ester groups [46]. The thermal properties of PET are:

- T_g: 70 to 115°C.
- T_m: 255 to 270°C [10].

This cohesion between PET molecules is also responsible of good mechanical properties, presented in Table 4.

E (GPa)	UTS (MPa)	Elongation (%)	Impact strength ($J.cm^{-1}$)	OTR (barrer)	WVTR ($g.m^{-2}$)
1.7	50	4	9	0.02	18

Table 4: PET properties [18][43]

PET properties are quite different compared to other polymers presented in this paper. Its Young's modulus is the highest, and its low elongation demonstrates that a high crystallinity can be achieved with this polymer. The highest crystallinity among the polymers of this paper. Its low gas permeability is also an important property. It has an OTR of 0.02 barrer, which is six times lower than for polyvinyl chloride (0.12 barrer). Its CO_2 transmission rate is also really low, with 0.15 barrer, compared to 1 barrer for PVC [18].

Because of this high temperature and mechanical properties, PET can be difficult to process. In order to improve this point and obtain a more ductile material, while keeping these interesting properties which are thoughness and gas impermeability, copolymers are included in the used resin. Copolymerisation is a technique that consists in introducing different monomer units in the same chain molecule. It has the effect of combining properties of both monomers. By adjusting the percentage of the different units along the chains, properties of the final compound will be closer from one or another unit. In the case of PET, one of its most famous copolymer is PET-glycol modified (PETG).

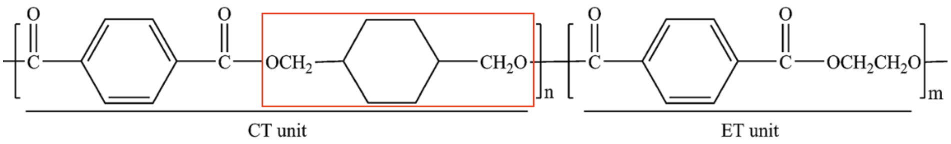


Figure 9: PETG structure, with cyclohexane dimethanol (red box) [47]

By using cyclohexane dimethanol instead of part of ethylene glycol, the structure presented in Figure 9 can be achieved. When the cyclohexane dimethanol unit content is about 40%, the PETG copolymer can be considered as amorphous [47]. This new copolymer keeps good mechanical properties, including impact strength and gas impermeability, in the same range of PET homopolymer. However, it has an increased transparency, and is easier to process because of its lower crystallinity and higher ductility. Manufacturers can modify the cyclohexane dimethanol content in order to achieve properties that suit their products requirements. For instance, for bottles production, a small percentage of co-monomer is sufficient to slow down the crystallinity and improve moulding of the resin, while keeping important properties for this sector.

2.4.3 Main applications

As explained in the above subsection about polyvinyl chloride, polyethylene terephthalate is a better alternative in terms of bottle packaging applications. Due to its higher gas impermeability, it allows to preserve the content and avoid gas exchange with the exterior environment. It is really useful, especially for gaseous content, including water and soda. For this reason, PVC bottles were drastically reduced. In the US, nearly all the juices, water or soda bottles are made with PET.

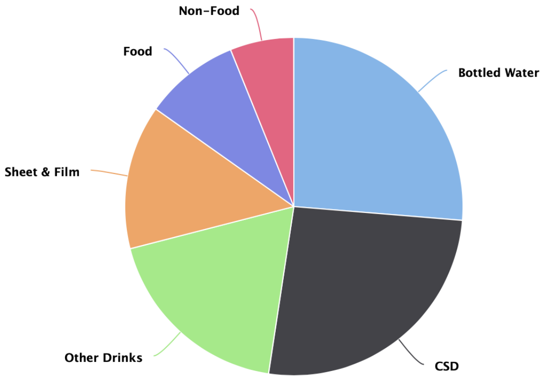


Figure 10: Distribution of PET materials worldwide (2016), from [48]

Figure 10 shows that almost 75% of the PET resin for PET materials production is used for bottles application. This figure only highlights the different sectors for PET materials, which does not include polyester fibres, used to produce clothes by replacing natural fibres like cotton. Other packaging applications for PET resin are, among others, films and food trays.

2.4.4 Products recyclability

PET bottles represent around 75% of the PET resin used for packaging, and more than 500 billion are produced each year. Hence, it is essential to increase their recycling rate. Moreover, these products, thanks to their volume and size, are less susceptible to leak in the environment, and have a higher chance to be collected for recycling. They are characteristic among plastic packaging and deposit system for their collection are often implanted.

In the case of PET, the most problematic associated material in packaging is PVC, used with PET as seals or labels. As described in the above content, PET starts to melt at a temperature around 260 °C. At this temperature, PVC decomposition occurs, and it engenders the formation of HCl. This compound can be a real contaminant for the recycled content. It has an impact on mechanical properties of recovered PET and its transparency [49]. It is also complicate to separate PVC and PET before melting process as their density is close. Polymers associated with PET should have a lower density to facilitate the sorting. As for other polymers, coloured and opaque PET has a lower recyclability and is less interesting for the recycled content market because of a lower range of applications.

The main use of recycled PET is in the textile sector. Until recently, recycled polyethylene terephthalate could not be used for food-contact packaging or bottles, because of bad smell and color. With the development of recycling methods, especially chemical recycling (cf. 5.3), a recycled PET with higher quality can be obtained. Smell problem has been solved and transparency is acceptable.

Figure 11: PET bottles made with different degrees of recycled content [50]

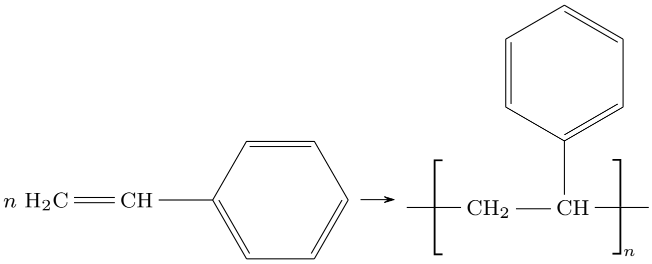
The bottle still turns into a yellow pale color, but it is acceptable for commercial application (Figure 11). Moreover, companies created additives that allow to eliminate this yellow shade with recycled content, but their use needs to be kept under special conditions. The additives needs to avoid any bad effect on the recycling process, because PET bottles made of recycled content can still be recycled up to several times. When it cannot be used for bottles production anymore, PET resin is used to make fibres and feeds the textile sector.

2.5 Polystyrene (PS)

2.5.1 Structure and reaction process

Polystyrene, and its expanded form particularly, is maybe one of the most distinctive plastic of the packaging sector. Even if it was developed during the 20^{th} century mainly because of its important electrical properties (electrical insulation), this polymer has other interesting properties which make it one of the most used resin in the packaging sector. In 2018, its produced volume worldwide was around 10 Mt and it is supposed to increase in the future, as for many other types of plastics. In 1999, in the European plastic packaging sector, polystyrene accounted for 7% of the market, and expanded polystyrene (EPS) for 2% [9].

PS is made by polymerization of ethenylbenzene monomers, more commonly called styrene, whose chemical formula is C_8H_8. Even if, as for polypropylene and polyethylene, PS monomer is only constituted of carbon and hydrogen atoms, it is included in the polyolefin family, because the styrene formula does not respect the $\mathrm{C_nH_{2n}}$ criterion. The main process for PS polymerisation is bulk polymerisation. It consists in adding monomers and radical initiator, creating a mix liquid solution. The reaction is activated by heat. The temperature for the reaction is in the range 100 to 200°C, and after formation of polymers, volatile monomers are removed in another step at a temperature around 200°C [51]. This bulk polymerisation is exothermic, which can be a problem during the formation of polystyrene. Because both monomer and polymer are bad thermal conductors, it is difficult to evacuate the engendered heat of the reaction and it could become uncontrollable. This heat generation has to be well monitored to keep this polymerisation process under control.



Polymerisation of styrene monomers

As for polypropylene, tacticity is present and influence the nature of the polymer. The phenyl groups can be located on the same side of the main chain (isotactic form), alternatively positioned along this chain (syndiotactic form), or randomly placed (atactic form). An illustration of these three different forms can be found in 2.3.1 above. The more the polymer is under isotactic form, the more the structure is ordered, and the higher the crystallinity of the polymer. Commercial polystyrene is amorphous and atactic. Because phenyl groups are not aligned on the same side of the chain, and that the layout is not ordered, it has an amorphous behavior. However, compared to polypropylene, polystyrene owns a phenyl group in its pattern structure. This group occupies a bigger volume than a methyl one and its movement is made complicate. The presence of these unwieldy and bulky groups along the chain implies a high rigidity, and high temperatures are required to make the chains move and bend. Even if atactic PS is considered as amorphous, phenyl groups size increases its glass transition temperature compared to PP. Syndiotactic and isotactic forms of PS can be processed, but their commerical interest is lower. Currently, commercial PS is under atactic layout.

Density of commercial PS is around 1.05 $g.m^{-3}$. Its number average molecular weight $\overline{M_n}$ is in between 100,000 and 500,000 depending on the degree of polymerisation achieved for the final polymer [10].

2.5.2 Properties

As explained above, due to the phenyl groups and atactic layout, polystyrene is considered as amorphous, but its properties are close to polymers with good crystallinity, including PET. Because it is amorphous, PS does not have a melting temperature, only a glass transition temperature. However, this temperature is relatively high: T_g is around 100°C, but several tests showed that it can reach 107°C [52].
This temperature is different from syndiotactic and isotactic forms. Due to their ordered layout, these two forms have a higher crystallinity and are not considered as amorphous. Hence, they have both glass transition and melting temperatures [53]:

- Syndiotactic: T_g is around 100°C and T_m around 270 °C.
- Isotactic: T_g is around 100°C and T_m around 240 °C.

Apart from T_g and T_m, properties of syndiotactic and isotactic forms will not be presented in this report. Table 5 presents properties of commercial PS (atactic form, amorphous):

E (GPa)	UTS (MPa)	Elongation (%)	Impact strength ($J.cm^{-1}$)	OTR (barrer)	WVTR ($g.m^{-2}$)
3.3	50	2	0.2	1.1	>1000

Table 5: PS properties [10][18][54]

Basic form of polystyrene, known as general purpose polystyrene (GPPS) in the commercial sector, whose properties are highlighted in Table 5, is a transparent and brittle material [54]. It has a low impact strength and a poor water vapour impermeability, the poorest among the different polymers presented in this paper. But polystyrene is also known for its expanded form, a polystyrene foam called expanded polystyrene (EPS). It is maybe the most recognizable form of PS. In the common language, the word *polystyrene* is often used to describe this foamed form of polystyrene.

E (MPa)	UTS (kPa)	Elongation (%)
10	80	5-13

Table 6: EPS properties [55]

With pellets of PS resin, an added compounds allows the formation of a foam. This compound is then evaporated because of increasing temperature and expanded polystyrene foam is obtained. The final product has a low density (0.02 $g.cm^{-3}$) and is composed at 98% of air [56]. Its properties (Table 6) and aspect are different than GPPS ones. Despite the several properties highlighted in the above table, EPS has several interesting properties. In this form, PS has a better thermal insulation, water vapor resistance and impact absorption compared to GPPS [57].
A third form of polystyrene is found on the market, high-impact polystyrene (HIPS). As its name suggests, the main interesting properties of this PS form is its high impact strength. Difference between the two other PS types is that this high impact PS

implies a second polymer for its formation. A polymer chain, generally polybutadiene, is used as a frame for the final compound [58]. Then, PS chains are fixed along this polybutadiene frame, in order to create a polymers mixture (Figure 12).

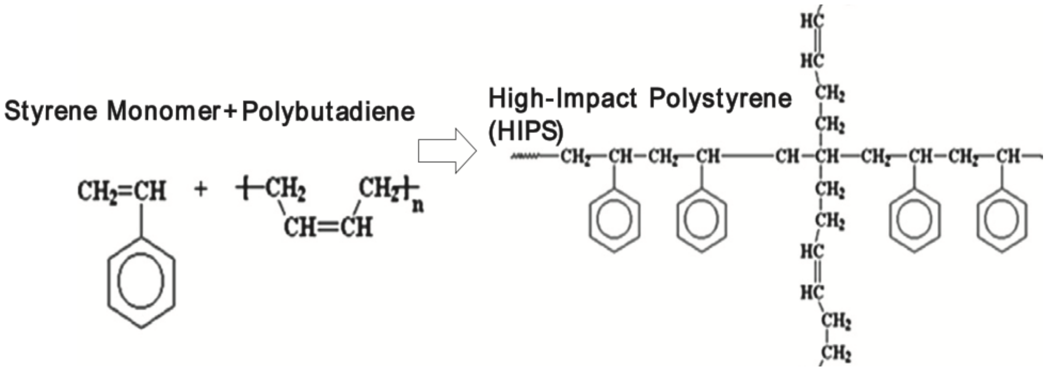


Figure 12: HIPS structure [59]

However, polybutadiene represents only a small percentage of the final compound, around 7% in average. This modification, though, allows to obtain a higher impact strength compared to the two precedent forms of PS. HIPS properties are presented in Table 7.

E (GPa)	UTS (MPa)	Elongation (%)	Impact strength ($J.cm^{-1}$)
2	80	10-50	3.5

Table 7: HIPS properties [60][61]

Moreover, products made with HIPS can be transparent on top of getting a higher impact strength. GPPS, EPS and HIPS are the three main types of commercial polystyrene, each one owning different properties, useful for different uses.

2.5.3 Main applications

Depending on their requirements, packaging suppliers will be able to choose between the different types of polystyrene presented above. The main packaging applications for polystyrene, according to the different types, are described below.

- GPPS and HIPS: cases (CD, video games, etc.), yoghurt and drink cups (including famous white and red party cups), food trays/containers and plastic cutlery.
- EPS: protecting packaging, take-away clamshells for food and hot content cups.

GPPS and HIPS applications were not differentiated because both materials can be used for the same products. The final choice only depends on the suppliers. However, GPPS will be used in general for cases and food trays/containers, whereas HIPS will be used for cups and cutlery [62].

2.5.4 Products recyclability

The majority of the PS packaging waste is composed of EPS. Indeed, it is due to its short-life applications, compared with HIPS and GPPS. Nearly all types of polystyrene, including HIPS, EPS and GPPS, can be recycled. In fact, on paper, it is even 100% recyclable. However, as for other polymers, some problems exist; and in order to recycle wastes engendered by PS products, some conditions must be respected.

Indeed, when labels are used with PS products, it is better for recycling if they are made with PET. In this case, a mix of PS product and PET label is not a problem. Because of the densities difference in between the two polymers, and also the high melting temperature of the polymer, a good recycling rate is possible for these products [63].

In fact, one of the major obstacle to polystyrene recycling, and especially EPS, is the poor ratio weight over volume [64]. Indeed, because of its high air content and high volume required for goods protection, EPS waste transport will require a higher capacity, compared to other polymers, for the same weight. The recycling process is facing the same problem. For this reason, most recyclers do not accept it in their recycling plants. Another obstacle is food contamination. As presented in the above part, EPS and PS in general are used for food trays, especially fresh meat or fish. In order to be recycled, they need to be rinsed before. But even after rinse, the recovered polymers cannot be used to make new trays or plates for food, due to the potential risks for human health.

2.6 Packaging polymers comparison

Graphs comparing the different properties of the polymers presented above have been placed in the Annex section of this paper.

When choosing a material for their product packaging, brands have to define precisely the requirements in order to protect the content of the package. Few criteria can be more important than others, and concessions can be made in order to avoid useless costs. Nowadays, the main criterion for polymer type selection are content preservation and packaging cost. The recycling criterion is still not considered as fundamental among companies. Similarities in the recycling conditions exist between the different polymers. For instance, contamination by other polymers has to be reduced at most. But all the plastics have not the same recycling rate. The most recycled, among the polymers presented, are PE and PET. PET is mainly used for bottles in the packaging sector, and these packages have quite a low degree of contamination by content and a high collection rate. However, regarding other polymers, recycling process and rate need to be improved. Sometimes, the nature of the polymer is responsible, as for PVC and its high content in additives that complicates the recycling. In this case, replacement of the problematic polymer by another one could be more efficient, easier and quicker than developing new recycling processes.

The objective of this part was to present and develop the main polymer types used in the packaging sector and their specificities. Now, this paper will focus on the waste engendered by the plastic packaging sector, and how entities deal with these products reaching their end-of-use.

3 End-of-use treatment

In Europe, in 2017, more than 75 Mt of packaging wastes were created. Among this amount, plastic packaging wastes accounted for 20%, which represented around 15 Mt [65]. This percentage can be considered as low compared to the 40% of paper and cardboard packaging wastes, but the recycling rate is far lower concerning plastics. Most of the paper and cardboard packaging wastes comes from transport application, or external protection, and has a low contamination degree. Moreover, paper recycling rate is nearly reaching 100% in many developed countries.

3.1 Collection and sorting

3.1.1 Waste producers and collection

Among plastic packaging waste, two main categories can be highlighted:

- Waste created by households, known as municipal solid waste (MSW).
- Waste generated by industrial sectors, from factories.

The second category is not the main problem to deal with. Most factories have implanted collection systems with waste companies, to facilitate the waste management. Their wastes are collected in big volumes, often already sorted. Moreover, most of the waste engendered is not contaminated, which makes its processing and recycling easier. It is often large amounts of protective packaging, including plastic wraps or cardboard. Due to their nature and relatively good state, these wastes are not the main problem regarding waste management [66].

However, MSW can be a real problem depending on the collection measures implanted on the spot. Whereas plastics are nearly the same all around the world, the collection methods are sometimes different between cities of the same country. Thus, three main collection types can be highlighted for plastic packaging wastes [42]:

1) Plastic packaging wastes are collected in a special container. Each type of plastic has its own bin, or different types are placed in the same bin, as a mix of plastic packaging wastes.
2) A recyclable bin allows to collect different types of recyclable wastes, including plastics, cardboard, metals and even glass.
3) All the household wastes are collected in the same bin, from rubbish to recyclable ones.

Assuming that for each collection type, the same recycling rate is targeted, it means that a choice needs to be made between households' or government/private sector sorting.

Type 1) seems to be the best option, because the risks of contamination of recycling stream by other materials is lower, but it totally depends on the efficiency, the implication and the education of the population regarding sorting process. In most cases, especially North and East Europe, this collection is implanted for specific types of plastic packaging wastes, often PET bottles, with sometimes a deposit system. This deposit system has a significant impact on the recycling rate for plastic bottles [67], but is again depending on the population. Whereas deposit system was first implanted by companies in order to reduce the packaging costs by reusing them, it is now often managed by governments, which decide of the deposit amount. This amount is financed by companies taxes, as part of the Extended Producer Responsibility (EPR, cf. 4.4.1). By increasing the deposit amount, positive impact on recycling rate can be achieved [68].

Type 2) is maybe the most common option encountered today. Governments implant general "recyclable materials" bins and sorting is done after collection, by professionals and specialized infrastructures. In France, in order to facilitate sorting for households, since 2019, all plastic wastes can be placed in the same bin with metal packages, cardboard, paper and plastic bottles. Before 2019, only plastic bottles and flasks could have been disposed in this bin [69].

Type 3) is present in countries where recycling is not well implanted, and where nearly all wastes are disposed in landfills or incinerated.

The type of system used in a country is linked to its economical development, its recycling and sorting infrastructures and households' education regarding sorting and how they pay attention to it. For instance, in Japan, there is no deposit system for plastic bottles, however the material recycling rate (84,8% in 2017) is equivalent to Sweden one (84%), where a deposit system is in place. Collection systems are drastically different between these two countries. While in Japan, there is mainly a kerbside waste collection, Swedish have to bring their wastes to collection points [70]. Collecting system implanted is really dependant on the population. Thus, the current Japanese system works in Japan because its population is respectful of these rules, which are complicate to respect for foreigners (Figure 13).

- Bunches (up to 30 in diameter by 50 cm in length)
- Thickness, Up to 10 cm thick

- Put out dirty plastic containers or packages (with the plastic mark) as combustibles.

* Always be sure to empty spray cans, gas cassettes, and cigarette lighters, and place them in a transparent bag separate from other non-combustible garbage. Never attempt to puncture items listed above to avoid a dangerous situation.

Sort recyclables by item and place in a transparent bag or recyclables collection net.

This refers to plastic containers, bags, and packaging containing the products, that have the plastic mark.

- Always be sure to empty all content and wash off any dirt before putting out. Put out dirty recyclables as combustibles.

Figure 13: Households' sorting rules (Japan)

Once sorting has been done by households, plastic packaging wastes need to be transported to sorting and recycling or disposable factories. In developed countries, waste transport is done by municipalities or private companies under contract. The allocated budget for wastes collection and transport can reach 50% of the budget for the total waste management. In developing countries, this collection is still partly realised by the informal sector and waste pickers. This phenomenon is well known in India, where around 1.5 million of waste pickers were reported in 2010 [71]. Only a small percentage of these waste pickers is supported by local authorities. However, their work is non-negligible regarding MSW management. In Delhi or Pune, two cities whose population is higher than 3 million inhabitants, this informal activity represents around 20% of the recyclable wastes collection. In this sector, it is one of the main challenge for India during the next few years: to improve its collecting process without removing waste pickers' job and source of income.

3.1.2 Sorting process

After being collected, plastic packaging wastes are pre-sorted by municipalities or collecting organisms, before being sent to companies whose only activity is wastes sorting. These plants can be the same than recycling plants but it is not frequent. Generally, these companies sort wastes before sending them to recyclers. Ariake Kogyo factory, in Koto city (Tokyo), receives pre-sorted wastes from Tokyo municipalities and sort them to produce bales of plastic and metal packaging wastes (Figure 14). These bales are then sent to recycling factories. Ariake Kogyo is also producer of burnable material from plastic and paper wastes, which do not suit recycling criteria. This burnable material is used by industrial companies in replacement of coal in energy production.

Figure 14: Mix of plastic wastes (left) and plastic packaging bales to send to recyclers (right) in Ariake Kogyo factory (Koto city)

Once in recycling factories, another sorting is realized, more specialized into plastic types. It is the case of the Veolia factory in Saitama (Japan), which receives bales of plastic packaging pre-sorted (Figure 15). This method is essential to limit contamination of the recycling stream. Sorting allows to separate different types of plastic, but also other materials part of the recycling process, and which can be collected with plastic packaging, including glass, metal and even paper. Sorting methods can be used to separate directly the different types of plastics (source separation), but also to focus on one type precisely, in order to eliminate it from the stream. In this case, other sorting methods are used to separate wanted plastic types after.

Figure 15: Bundles of pre-sorted plastic wastes arriving at Veolia plant (Saitama, Japan)

Depending on the collection method, different sorting systems can be applied:

- Manual sorting is still used in factories, even if main part of the process is automatized nowadays. Operators can be upstream of the sorting process, in order to realize a coarse sorting at first, or downstream, to do a quality check. However, an effective sorting cannot be based only on manual sorting due to the higher probability of inaccuracy and the huge quantity of waste to process [72].
- Magnetic sorting allows to separate plastic and metal contaminants, to separate both ferrous and non-ferrous metals from plastic wastes, with three different paths.
- Density sorting, which can be realized in solution or by air stream. Due to their different densities, plastic types will not react in the same way to the fluids.
- Electrostatic sorting uses an electric field through which plastic wastes are sent. Depending on their electrostatic charges, their behavior will not be the same when submitted to this electrical field, and they are separate thanks to this difference [73].
- Optical sorting regroups near infrared (NIR), X-rays and color methods. Their principle is based on the response of wastes to waves of different lengths

> (infrared, X-rays or visible domain). While NIR allows to sort at high velocity many types of polymers, including products made with polylactic acid (PLA), with a low inaccuracy, X-rays sorting is mainly used for the detection of a special polymer, polyvinyl chloride, whose response to these waves is drastically different than for other polymers.

NIR is one of the most viable sorting options for plastic wastes mix. It uses IR waves, whose wavelengths go from 750 to 2500 nm approximately. In this range, polymers used for plastic products absorb differently these waves, and an analyze of the IR absorption spectrum allows to discover the nature of the polymer (Figure 16).

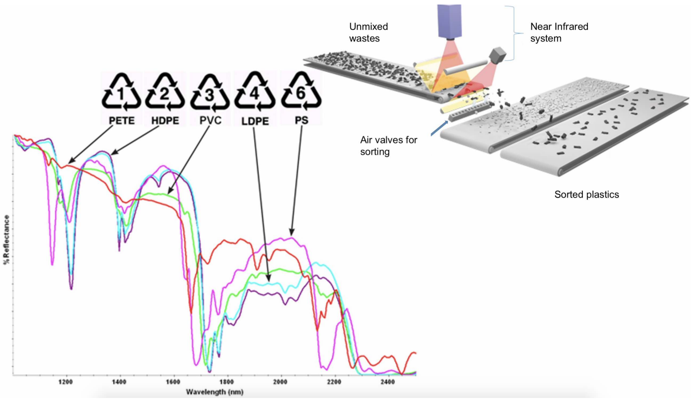


Figure 16: IR absorption of different polymer types in NIR sorting process [74]

By using NIR detectors, the sorting chain analyzes at high speed the constitution of plastic wastes (Figure 17). It uses air valves to drive and sort the different plastic types in the dedicated containers for being recycled.

Figure 17: Near Infrared sorting machine at Veolia plant. Upstream (left) and downstream (right) of the sorting process

All the systems described above are often used as complementary methods for plastic wastes sorting. New systems are currently developed to improve sorting efficiency and avoid contamination of streams. Tomra Sorting Recycling, a company creating sorting machines to improve recycling, plan to commercialize a sorting system using a robotic arm, sensors and artificial intelligence. Through deep learning, this system can select different types of plastic packaging, from film to heavier products, but also paper [75]. In order to help for sorting, each resin type can be identified thanks to its own code (Figure 18). This identification code allows plastic packaging sorting and recycling by precising explicitly the nature of the resin used for a product. It is particularly useful for manual sorting and operators.

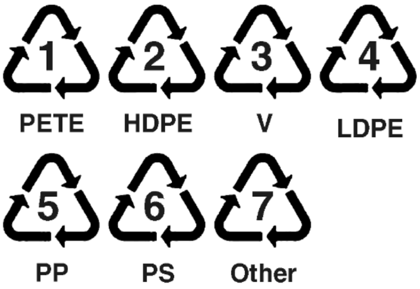


Figure 18: Resin identification codes [76]

These identification codes were created at first in order to facilitate detection of bottle packaging made with recyclable plastic. The number were given type by type, progressively, when recycling of the resin was discovered and democratized. In the common imaginary, these symbols, and especially the triangle with arrows, indicate that the material and package is recyclable, but it is not true. Types 1 and 2 are frequently accepted for recycling, whereas others are not, especially codes 3, 6 and 7 [77]. Reasons for poor recycling of certain types of resin were detailed in the previous section.

The next parts will describe the treatment of plastic packaging wastes after being sorted.

3.2 Landfill and incineration

Landfill and incineration are the less profitable regarding the circular economy, because they do not produce any material or energy from the plastic packaging wastes. It is important to differentiate incineration from thermal recovery, which will be developed below. On one hand, incineration plants only eliminate waste, to avoid waste flooding. On the other hand, thermal recovery is also a process that applies a thermal treatment to waste, but these plants produce energy from this thermal treatment, by using calorific value of waste. For this reason, the process is often called thermal recovery, Energy-from-Waste (EfW) or Waste-to-Energy (WtE). Incineration without energy represents only a low percentage of the current incineration plants, around 1% [78]. However, all the EfW plants have not the same energy yield or impact on population. More sophisticated plants, located in developed countries, with security improved regarding health concerns, are different from plants in developing countries, which have a real impact on the surrounding population.

Landfill is another problem faced by waste management nowadays. In areas in development, it is still the main way to manage wastes. Around 70% of wastes in general are placed in landfills in the world. This percentage reaches 90% in some areas, including Africa [78]. This high percentage is due to the lack of money and development concerning recycling. But regarding plastic packaging wastes, the percentage sent in landfill is high even in developed countries, often due to recycling difficulties with these materials and the lack of infrastructures. In 2017, in the US, around 14 Mt of plastic packaging wastes were produced, from which 70% were sent to landfill [79]. In 2019, in Japan, 6% of post-consumer packaging wastes were sent to landfills and 12% incinerated without energy production [80]. Landfilling of plastic packaging is dangerous for environment because it is not a material that will degrade easily. Moreover, as they are lightweight, these packages are more likely to leak into the environment, especially with open sky landfills. One other problem linked to landfill is the methane and carbon dioxyde production. Mixing of wastes engenders production of these gases, which are greenhouse gases. In the US, wastes landfilling is the third activity linked to human in terms of methane production.

However, more and more landfill plants collect gases, especially methane. By this process, this global warming gas is not released in the atmosphere and can be used for different activities. The main uses of collected methane are electricity production, direct use (user is connected to landfill via a pipeline and burns the gas for its activities) or refining to produce higher quality gas [81]. These landfills using methane production represent around 560 plants in the US (2020) and 500 others could join. Development of this type of landfills is important, because it allows to drastically reduce the amount of gas released in the atmosphere. In the US, each year, around 2 Mt of methane are collected thanks to these plants [82].

3.3 Compost

3.3.1 Process

Composting is different than landfilling because it only manages organics wastes and the gas produced during degradation is mainly CO_2. The absence of methane production (or low) is due to the degradation process. Indeed, composting uses oxygen through an active degradation [83]. In order to submit compost feedstock to oxygen action and aeration, factories can use machines to stir it, or place ventilation under and force air flow through it, as for aerated static composting. All these parameters allow proliferation of microorganisms that degrade wastes, through an active degradation (Figure 19). Moreover, composting provides a useful material at the end of the process, compost, which can be used for agriculture in order to reduce the use of chemical substances of this sector. There is an important difference between home composting, that households can realize and which requires only stirring and industrial composting. Industrial composting facilities manipulate several parameters, in order to modify wastes environment, increase degradation and control gas emissions. The temperatures in these plants are varying between 50 and 60°C to accelerate degradation process and eradicate hazardous particles [85]. Humidity and oxygen levels are also under control. From now, when using the term *composting*, this paper will refer to industrial composting.

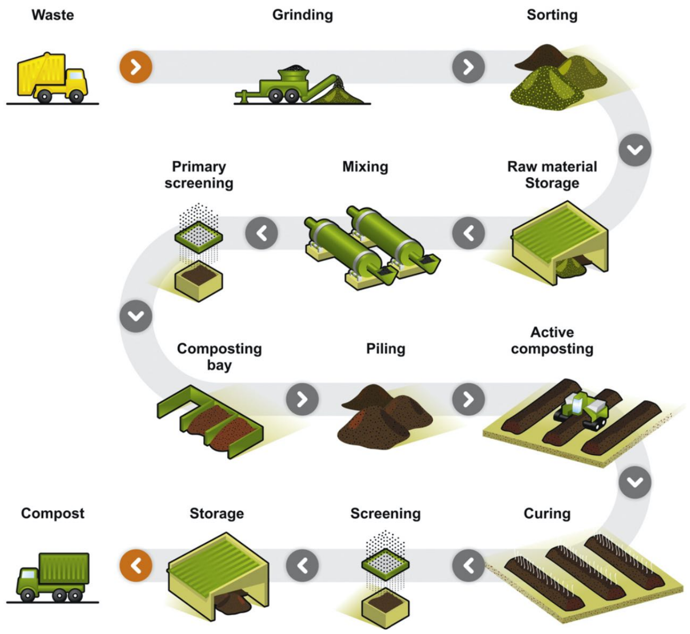
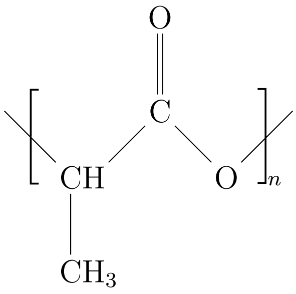


Figure 19: Industrial composting process [84]

3.3.2 Polylactic acid

Classic synthetic polymers depicted in the first section cannot be processed through composting. Their process of degradation in this environment, even with modified conditions, is too long. However, several types of bioplastics, made with bio-based content, can be biodegraded in compost factories. It is important to notice that all the bioplastics cannot biodegrade. The development of bioplastics is not recent. Indeed, as introduced above, the first plastic created was a bio-based plastic made with cellulose [86]. The development of these plastics was slowed down in the 20^{th} century, due to petrobased plastics low price and better properties. In the past decades, new bioplastics were developed with improved properties, leading to a restored interest for these plastics. One of the most famous bio-based and biodegradable plastic is polylactic acid (PLA) plastic.



Polylactic acid structure

PLA is made with bio-based resources, including corn and sugar cane, and it was first developed for the medical sector, its natural degradation being interesting for implants. Now, due to countries ban of petrobased plastic bags, one of its main applications is grocery bags. PLA properties are close to several petrobased polymers, but it has weak points, especially OTR, WVTR and impact strength [87]. Recently, these properties have been improved through different methods, including mixing with other polymers and additives. It is able now to meet expectations of the packaging sector.

Degradation of this material is easier than for classic polymers, which is the key aspect of its development (Figure 20). There are two main parameters that can initiate degradation process of PLA: water and temperature. Water is the initiator of thermal degradation and has an effect on the ester group of PLA. Indeed, the water molecule will react with ester, leading to the formation of carboxylic acids and hydroxyl groups. This mechanism is called ester hydrolysis:

$$R_1{-}COO{-}R_2 + H_2O \longrightarrow R_1{-}COOH + HO{-}R_2$$

In fact, both degradation processes have an impact on each other: temperatures can accelerate this water degradation (even for $T < 60$°C) and, during PLA processing at high temperatures, hydrolysis is the precursor of thermal degradation [84]. Both thermal and hydrolysis degradation processes are important for the composting of PLA-based products. Indeed, this polymer can be recycled as others, but due to the low proportion of this material on the market, there is a limited number of factories which are dedicated to its material recycling and it is considered as non recycled. Then, the most interesting waste management process is composting. Because these composting plants use water (high humidity) and heat, PLA degradation is efficient.

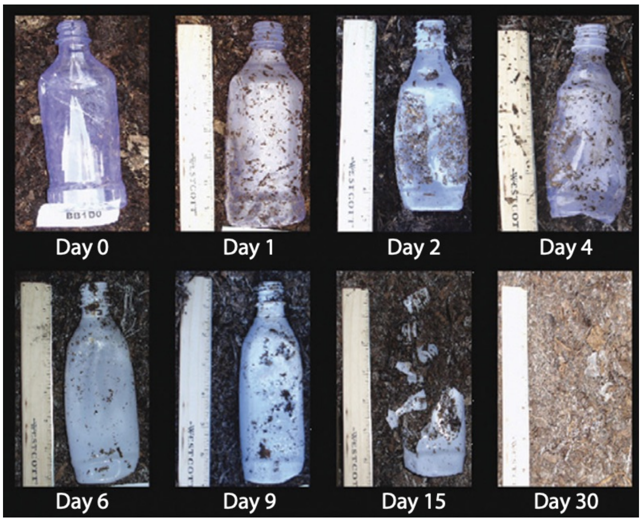


Figure 20: PLA composting results ($T = 58$°C and modified humidity) for bottle packaging [87]

PLA-based packages could be interesting to replace petrobased packages in the food sector. Because these packages are contaminated, their material recycling is complicate. This problem is not present through composting, where food contamination can even play a precursor role in the development of microorganisms. At the moment, PLA products composting, and biodegradable plastics in general, is facing two main problems in its development.

First is the difficulty to sort these materials and differentiate them from petrobased polymers. Thus, a large portion of composting factories does not accept PLA for compost, whereas it is its dedicated path. This bioplastic ends up in landfill, burnt or in other polymers recycling streams, where it acts as contaminant. Second, the effect of this polymer expansion on resources is not quantified yet. Production of corn and sugar cane requires space for fields, which could lead to an increase of deforestation in sensitive areas.

PLA and other biodegradable plastics could be an opportunity to replace existing materials, in contaminated and lightweight packages, and improve wastes processing through compost. However, improvements are required for sorting process. One dedicated application for PLA products could be in closed-circuit of events, including concerts, conferences or festivals. During these events, people have no other choice than buying products on the spot and using bins that staff makes available. Thus, it could be possible to collect only PLA wastes, in large quantities, and use a transporter to send these wastes directly to compost facilities, while avoiding contamination. Indeed, it is frequent for events to use only biodegradable packages.

3.4 Thermal recovery or Energy-from-Waste

The concept of Energy-from-Waste was already introduced above with incineration without energy production (cf. 3.2). However, there is not a simple and binary difference between these two processes. Around 80% of EfW plants are located in developed countries, where concerns exist regarding emissions and residual wastes management. Often, as in Europe, directives have been implanted in order to regulate and oversee these plants development [88]. Thus, effect on surrounding population will not be the same between an EfW plant in a developed country and in a developing one.

When composting is not possible, as it is the case for many plastic packaging actually, or material recycling, because of contamination or other difficulties, thermal recovery emerged as one of the best options. It allows the production of energy and decreases the amount of wastes. There is no doubt that diversion of these non-recycled plastic wastes from landfill to thermal recovery could have a positive and ecological impact. For instance, in the US, the non-recycled plastic wastes placed in landfills represent around 80% of the total amount of these wastes [89]. In 2017, 17% of plastic packages and containers were sent to EfW plants in this country. In Japan, in 2019, 59% of the post-consumer plastic packaging were processed through thermal recovery plants [80].

3.4.1 Heat value

One important notion when presenting EfW process is the heat value (HV) of wastes. This value describes the quantity of heat discharged by a material during its combustion. The unit of these property is given in $J.kg^{-1}$. The higher the HV, the

higher the energy produced by the material during combustion.
HV can be separated into two subcategories: high and low HV. The difference between both notions is that, in the case of high HV, heat generated by water condensation at the end of combustion process is taken into account, whereas it is not the case for low HV. These two notions are linked as follows:

$$[high\ HV] = [low\ HV] + [water\ condensation\ heat]$$

In commercial EfW plants, water steam is evacuated with other gases. Thus, heat generated by water condensation is low, and it is more interesting to focus on low HV. Moreover, both values are close in the case of non-recycled plastic wastes. Table 8 highlights low HV for common non-recycled plastic wastes.

Plastic type	PET	HDPE	PVC	LDPE	PP	PS
Low HV ($MJ.kg^{-1}$)	23.8	37.1	22.9	42.6	41.0	38.6

Table 8: Non-recycled plastic wastes low HV [89]

According to this table, the plastic waste type creating the highest amount of heat during combustion is LDPE and the lowest amount is PVC. In general, PS and PVC are known as non-recycled plastics, due to additives quantity and diversity for the first one, and low ratio mass/volume for the second (especially EPS). EPS has also a low HV higher than 40 $MJ.kg^{-1}$. While material recycling is not interesting for EPS wastes because of high transport costs, linked to its low density, and contamination, they have a real EfW interest. Those HV are higher or comparable to coal low HV (around 25 $MJ.kg^{-1}$). Moreover, the lowest HV accepted for thermal recovery, which corresponds to the heat value necessary for maintaining combustion, is 7 $MJ.kg^{-1}$ [90]. Thus, thermal recovery of non-recycled plastic wastes, and in particular plastic packaging wastes, is a good opportunity to create energy and realize wastes diversion from landfills.

3.4.2 Process

Most EfW plants use mass burn process for energy production. These factories use a stocker, in order to stock a large amount of mixed wastes, including plastic packaging ones. This stock will be consumed during several days, without interruption, and can be refilled at any time. In this part, the three main types of mass burn plants will be presented.
The most common is known as grid incinerator (Figure 21). Wastes from the stock will be sent on this grid system for combustion. Combustion takes place at temperatures higher than 1,000°C, in order to eliminate and consume efficiently wastes [91]. However, above 1,300°C, combustion could engender production of new hazardous gases and affect ashes constitution. The grid system is generally separated in three distinct grids. The first one, just after the stock exit, allows to dry the wastes. The second is where the main combustion takes place. The third one is for post-combustion, to be certain that all the burnable and useful content is incinerated

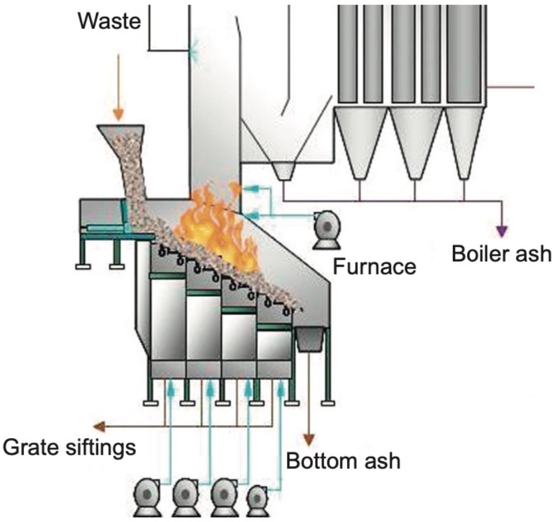


Figure 21: Grid incinerator process [92]

before eliminating bottom ashes. These ashes result from wastes combustion, and are managed through a dedicated track. It is the responsibility of plants operators to deal with these residual wastes, to avoid leakage in environment and atmosphere. Annually, in Wales and England, more than 500,000 tons of bottom ashes, produced by EfW plants, are disposed in landfills [92]. Recently, researches allowed to develop use of these ashes. Bottom ashes can be used for concrete production, with even a positive impact on mechanical properties of the produced material [93].
The two other types of mass burn process are less frequent than grid one. These are called fluidized-bed and gasification melting systems.

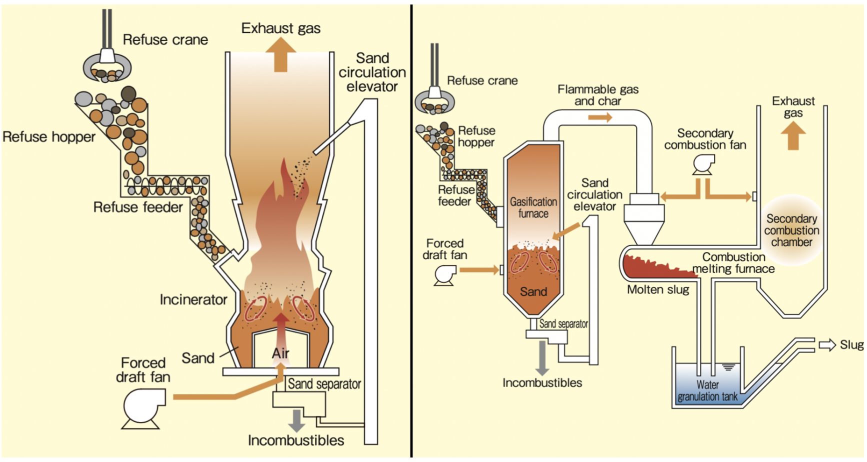


Figure 22: Fluidized-bed (left) and gasification melting (right) processes [80]

Fluidized-bed method, as depicted in Figure 22, does not use a dynamic but static system for waste combustion. From the stock, wastes are sent into an incinerator with a bed substance. Materials for this substance vary, but sand stays the most frequent choice. Under this sand bed, an air system is implanted. It projects air flow through the sand and, when air speed is sufficient, the bed substance gets a fluid behavior. This behavior is achieved with an air speed starting at 1 $m.s^{-1}$. If the speed continue to increase and reach 5 $m.s^{-1}$, bubbles are created in the sand in order to evacuate the excess of air [94]. Incombustible residuals are collected with sand at the bottom part of the incinerator, where they are sorted. Sand is then sent back as bed substance through the top of the incinerator. This process has several advantages and drawbacks compared to other incineration systems:

- Advantages: thanks to the fluid behavior of sand, combustion is more homogeneous, which prevents temperature (and so combustion efficiency) differences inside the incinerator; and lower temperatures (in the range 800-1,000°C) avoid hazardous gases formation.
- Drawbacks: wastes have to be shredded into pieces of homogeneous and small sizes before being mixed with sand.

Gasification melting process is different than the two others. Indeed, another step is added after waste burning, before the power generation part of the EfW plant (Figure 22). The first part is the same than for fluidized-bed method, wastes shredded in small pieces are introduced in fluidized sand. However, it is not a combustion but a gasification of these wastes. These gases are not yet used to produce electricity. Mixed with ashes and nonburnable particles, these combustible gases are introduced in the combustion furnace [95]. In the furnace, due to vortex flow and gas combustion, solid compounds are melted, cooled in contact with the wall surface, and finally fall at the bottom of the furnace for collection. The exhaust gas created by combustion is then used for energy production (Figure 23). This gasification melting process has several advantages. Thanks to gas combustion, hazardous particles created during gasification are eliminated. In addition, the melting of ashes create a recyclable material of higher quality than ashes collected at the bottom of sand bed.

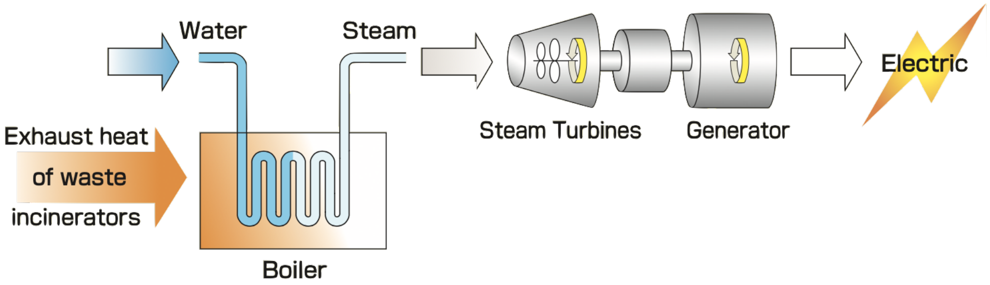


Figure 23: Power generation system [80]

For each of these processes, wastes or gas combustion engender formation of dissipate gases. These hot gases are used for electricity or heat production. Thanks to a boiler, water going through is boiled, producing water steam. This steam activates turbines rotation, linked to a generator, which creates electricity. Another type of EfW plants consists in using heat produced by steam directly, thanks to a heat exchanger. Water formed by steam condensation is then sent back in a closed loop system.

As explained above, part of residual wastes from EfW plants are bottom ashes, collected directly during combustion, and used for cement production or disposed in landfills. However, other hazardous particles have to be managed. These particles reach the end of the process with gases, and are considered as more dangerous than bottom ashes. These are flying ashes and volatile residues, including chlorides from PVC plastic wastes combustion. Flying ashes and other residues are grouped in a material category called Air Pollution Control Residues (APCR). Nowadays, these APCR are collected with filters and other devices in EfW plants. Then, they are often combined with wasted water, in order to form a solid mixture [96]. This mixture prevents the leakage of APCR in environment because of wind. It will then be sent to special landfills, dedicated for hazardous materials. Chemical and thermal treatments of APCR are under researches, but any of them is currently used at industrial scale.
Incineration of wastes started to grow in popularity after the 1950's. In particular, this phenomenon was observed in countries with restricted space, in order to replace landfilling. Those countries include Switzerland, Japan and others, whereas countries with important usable area did not follow the same tendency, including the US. Since the middle of the 20^{th} century, numerous studies have been done to prove the effect of emissions on population, through the increase of cancers, respiratory illnesses and others. However, no real exploitable and clear link between EfW plants and diseases increase have been proved [97]. Thanks to filters, hazardous particles emissions, including dioxins, can be largely decreased. Even if EfW process is often considered in statistics as thermal recycling, it is better to consider it as thermal recovering, because it does not participate to the development of a circular economy.

3.5 Material recycling

Compared to the precedent waste processing methods, material recycling participates to the creation of a circular economy regarding materials. It allows to reduce the quantity of virgin plastic used for new products, and reduces wastes quantity. In Japan, 22% of post-consumer plastic wastes are materially recycled [80]. In the US, 13% of plastic packaging and containers are reprocessed into usable material [79]. These rates are noticeably lower than the average among EU member states, which is 42% [98]. Nowadays, two main categories of material recycling can be highlighted: chemical and mechanical recycling. The first one is still in development, but is promising for the future, and recent progress and advances are described below. Concerning the second type, mechanical recycling, for both quantities and costs reasons, it is currently the main way to recycle plastic wastes efficiently. Indeed, in Europe, more than 99% of the recycled plastic wastes come from mechanical recycling [99].
Mechanical recycling occurs after sorting methods. Plastic wastes reach the recycling plant after being sorted, but sometimes the factory can realize a new sorting in order to increase purity and reduce at most contamination in the different streams. In the case of factories that realize both sorting and recycling, coordination between the process chains is easier. Pre-sorted plastic wastes are often prepared for mechanical recycling through the following steps [100]:

1) Shredding in small pieces.
2) Washing.
3) Density separation.
4) Drying.

Theses steps allow to obtain clean, sorted by types and dry pieces of plastic, ready for mechanical recycling. In Veolia plant (Saitama), NIR sorting method is used in order to eliminate PS wastes, which are not accepted for recycling. The materials collected downstream are PET, PP, PVC and PET. Due to their different densities, these plastics are then sorted by flotation process (density separation). In Saitama, only PP and PE are mechanically recycled. Sorted PVC and PET are sent to other recycling or thermal recovery plants. Mechanical recycling can process these plastic pieces into usable material. In order to describe this process, this paper will use pictures and data from low scale process, realized with Nihon Yuki company machines. Nihon Yuki manufactures machines for laboratory, to realize experiments on recycled plastics. These machines are called extruders, even if the word extruder refers to a part of the machine, where plastic wastes are melted. This is a reduced version of the recycling process used in commercial recycling plants, including Veolia plant in Saitama.

Figure 24: Recycling machine (left) and shredded plastic pieces (right)

After being introduced in the funnel (Figure 24), shredded pieces enter in the melting part of the machine, the extruder. This part is composed of a heated screw which melts and homogenize plastic particles (Figure 25). The extruder, and so the screw, has a temperature gradient along it. Generally, it is separated in three distinct parts, each one with a dedicated function:

1) Feed zone: introduction of the shredded pieces, which start to melt slowly due to the high temperature (T=85°C in this case).
2) Melting zone: temperature increases gradually in this part (temperature gradient) and the channel depth of the screw is decreasing, which implies melting acceleration (T=230°C at the end of this zone).

3) Metering zone: channel depth is low and constant, and temperature increases slightly. Aim of this zone is to melt last particles and mix the melted plastic into a homogeneous material (T=240°C).

The temperature gradient (∇T) in the melting zone can be defined as the variation of temperature per unit length. In the case of three dimensions dependant temperature, temperature gradient is the vector:

$$\nabla T = (\frac{\partial T}{\partial x}, \frac{\partial T}{\partial y}, \frac{\partial T}{\partial z})$$

If z is the axis along extruder screw, by supposing that the temperature depends only on this z axis and that the temperature diffusion is constant and homogeneous, temperature gradient norm can be defined as:

$$\|\nabla T\| = \frac{\Delta T_2}{L_2}$$

With ΔT_2 being the temperature difference between the beginning and the end of the melting zone, and L_2 the length of this melting zone. In the case of Nihon Yuki machine, it is possible to approximate the value of this gradient, knowing that $\Delta T_2 = 230 - 85 = 145K$ and $L_2 = 30cm$. Then, the value of the temperature gradient in the melting zone is: $\|\nabla T\| = 4.8K.cm^{-1}$.

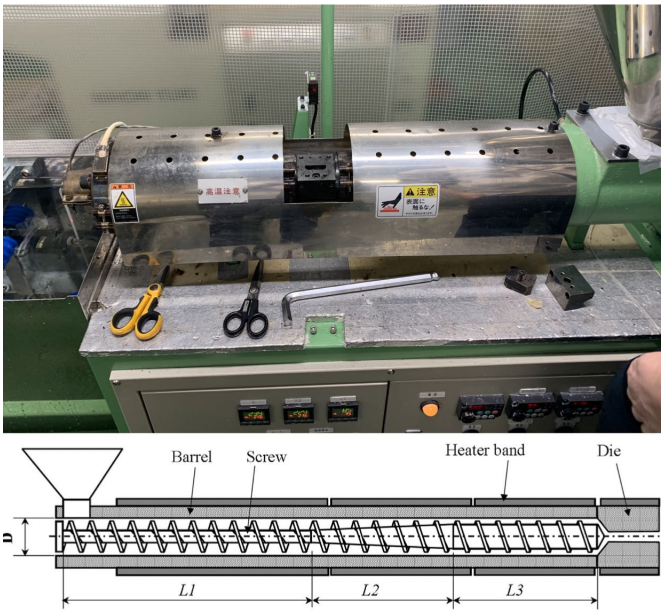


Figure 25: Extruder (top) and schematic version (bottom) [101]

Nihon Yuki machine screw has a diameter of 40mm and a rotation speed of 120 rpm. The temperature gradient is important for plastic wastes processing. If the temperature on the entrance of the extruder is higher than 200°C, polymers can degrade, loose properties and create a plug. Thus, friction and pressure inside the

extruder, which create extra heat, have to be taken into account. Pressure inside the extruder can exceed 30 MPa.
Depending on the rotation velocity of the screw, different phenomena can occur [101]. In the case of a too high velocity, it is impossible to obtain a homogeneous mixture, because plastic pieces do not have enough time to melt properly. When velocity is too low, polymer material encounters a too important melting, which has an impact on the form and properties of the final product.

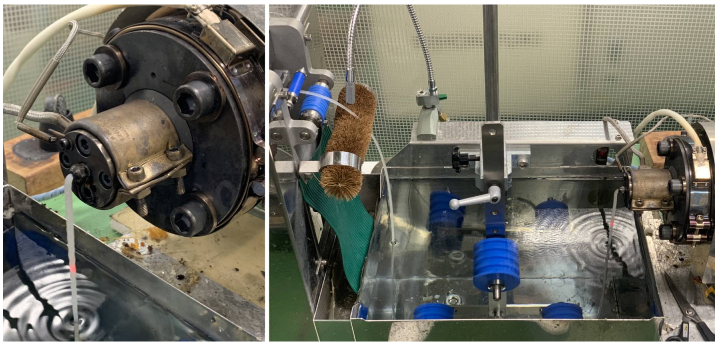


Figure 26: Extruder output (left) and wire cooling bath (right)

After being processed through the extruder, plastic material is shaped into a wire (Figure 26). Temperature of the wire is measured thanks to a laser sensor. The registered temperature is used in order to regulate the processing temperature inside the extruder. The plastic wire is cooled in a water bath, which turns it from soft to hard texture. An air sprayer at the end of the bath allows to dry the final wire, before being cut into pellets and collected (Figure 27).
The pellets production for Nihon Yuki machine is in between 6 and 11 $kg.h^{-1}$. This machine is used mainly for tests and researches. Veolia recycling plant in Saitama can process up to 36,600 tons per year (120 t per day) and, in 2019, the total recycled materials production was around 10,000 t.

Figure 27: Pellets production by Nihon Yuki machine (left) and Veolia plant (right)

The obtained material can be reprocessed into new products, but it has lost part of its properties and quality. Indeed, several challenges occur during mechanical recycling of plastic wastes, which compromise the quality of the new material.

The first challenge is the polymer degradation due to temperature, and external environment in general. The plastic wastes are first heated in the recycling plants, as seen above, but also in factories, in order to be reprocessed. This sequence of heating has an effect on molecular structure of the polymer. Due to O_2 presence, the chains of the polymers are degraded, which leads to a change of properties, especially properties at break and crystallinity. The deterioration type depends on the polymer type, as depicted in Figure 28. This degradation becomes more important after several reprocessing cycles, which highlights the fact that mechanical recycling is not a long-term solution for circular economy of plastics. Several additives, including heat stabilisers can be added to the recycled material, in order to offset the drawbacks engendered by recycling [102]. Moreover, during its use, plastic products have to support different conditions, more or less difficult, that can affect the properties and degrade the polymer.

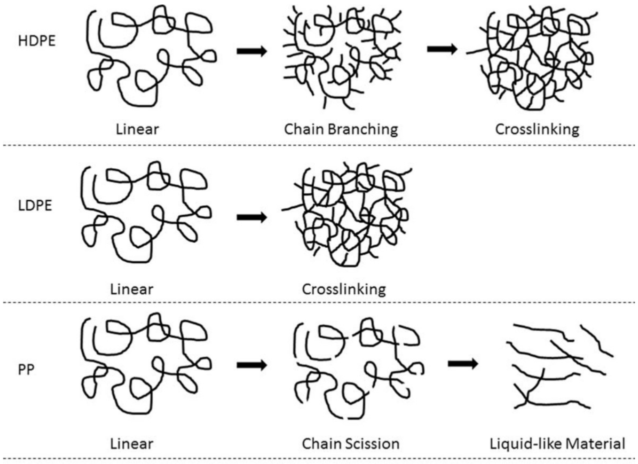


Figure 28: Oxygen and temperature degradation of polymers after reprocessing [102]

The second challenge is due to contamination during reprocessing. This contamination occurs because of other polymers, reaching the wrong recycling stream. As seen in first section of this paper, recycling of each polymer is complicated because of plastics association. Even with efficient sorting methods, including NIR sorting, contamination is inevitable. Indeed, NIR, as other optical sorting methods, analyzes plastic packaging surface. Some products being totally covered with shrink sleeve can be sent in the wrong stream. If PET bottle is totally covered with PE shrink sleeve, optical sorting will send it into the PE recycling stream, thus creating contamination. Stream contamination leads to polymers blend formation. This blend often encounters a mixing problem, due to the fact that polymers are non miscible. This is the case of isotactic polypropylene and polyethylene. This is problematic during melting steps, forming aggregates of different polymers [103]. Here too, additives can be used in order to make these polymers miscible, by increasing adhesion, but it affects purity and properties of the recovered material.

Current commercial recycling methods, through mechanical recycling, allow to obtain a recovered material usable for new products. However, there are several drawbacks with this method. Indeed, the recovered material is generally of lower quality, because of degradation and contamination by other polymers. It is really rare for a recovered material to be used for the same product than its original one, it is downgraded. This is especially the case for food or cosmetics plastic packaging. To make their recycled material usable for manufacturing of the same product, recycling companies have to present detailed report, certifying the quality and safety of the recycled plastic. Many places in the world, including European Union, implanted legislation regarding the use of recovered plastics in new products in direct contact with food [104]. Whereas it is difficult for plastic wastes coming from kerbside collection to be reused in these food-contact products, it is easier for closed loop products. PET bottles belong to this category, when they are collected separately at deposit points. Indeed, all the products of this plastic wastes batch are PET bottles, which were only in contact with liquid. Contamination risks are decreased, and it is easier for the recovered products to be used in new PET bottles. PET bottles collected through kerbside collection can also be reprocessed into new bottles, thanks to sorting, even if it is less efficient than with deposit collection. However, most of the plastic packaging wastes collected by kerbside collection has not the same recycling success. They are used to make plastic products with a longer lifetime, including furniture, pipes or textiles [105].

All the recovering processes described in this section have to be considered together and not separately. Mechanical recycling would not be possible if sorting methods were not efficient, and even these systems have their weaknesses. These weaknesses are compensated by collection and initial sorting from consumers. In order to improve continuously the end-of-use treatment of plastic packages, actions have to be taken by different entities linked to the packaging sector.

4 Entities involvement

This section will develop the different actions realized at the present time by the different entities linked to the plastic packaging sector. Ecology and environmental concerns are growing in the world. However, it is important to differentiate true involvement and biased one. Actions need to be thoughtful and not expeditiously taken, without knowing the possible harmful consequences for the future.

4.1 Nonprofit organization

A nonprofit organization (NPO) is known as an organization, whose only objective is to use its funding to assist the cause it defends. The causes are diverse, from animal rights to social and environmental developments. There is no benefit realized by NPO activities. Since several years, plastic packaging, and plastic in general, is known as one of the most problematic materials for both environment and human safety. Knowing this, several NPO started to focus on this problem, and other ones were created with plastic packaging sector improvement as their only purpose. However, these NPO are essentially located and created in Europe and North America (especially in the US). This part will highlight main NPO in the plastic packaging sector and their actions to upgrade it.

4.1.1 Ellen MacArthur Foundation (UK, 2010)

The Ellen MacArthur Foundation is not focusing only on plastic packaging, but on circular economy in general. Its vision is simple: with the growing worldwide population, the existing way of creating and consuming is not viable and will have a disastrous ecological impact. The foundation works in order to provide drastic changes in the current goods and wastes management.

Regarding plastic packaging problem, one of their main actions was the creation of the New Plastics Economy concept. This concept aims to develop a circular economy in the plastic packaging sector. It engendered the formulation of the Plastic Pact, soon joined by national and regional versions of this pact, and Global Commitments, signed by numerous companies. Every entity joining this New Plastics Economy movement, through Plastic Pact or Global Commitments, makes an engagement to achieve the Foundation objectives regarding plastic packaging sector. These objectives are:

- Increase the use of recycled content.
- Reduce the amount of plastic used in packaging.
- Develop end-of-use treatment of plastic packaging by increasing recycling and composting potential for these products.
- Avoid single-use (SU) packaging and focus on reusable options when it is pertinent.

The Ellen MacArthur Foundation gathers many companies and governments under this new economy, a circular economy. It led to the development of precise and concrete objectives announced by companies (cf. 4.2.1).

4.1.2 Sustainable Packaging Coalition (US, 2005)

This coalition was launched by another NPO, GreenBlue, and several companies from the packaging sector. The aim of this coalition is to make this sector sustainable by changing materials and also to create a circular economy. The coalition developed a guide to promote the use of recycled content in packaging, and more generally to improve the sustainability of used materials. But one of its main projects is the creation of the How2Recycle label (Figure 29).

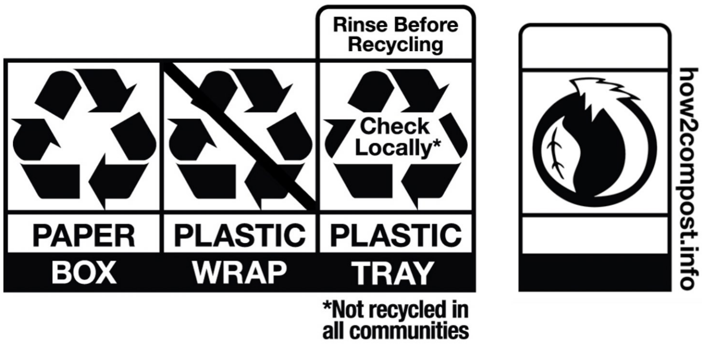


Figure 29: How2Recycle and How2Compost packaging labels

This label is the main one used on packaging in the US by many companies in the world. It provides instructions to consumer in order to increase sorting and recycling rate. The Sustainable Packaging Coalition also launched the How2Compost label, following the same principle than the How2Recycle one, but for compostable packaging.

4.1.3 Petcore Europe (Belgium, 1993)

The Petcore Europe NPO is specialized in plastic packaging using PET resin as main material. The objective of this organization is to promote the recycling of PET packaging and increase the use of recycled PET content. This improvement is possible thanks to the different groups created by the NPO. Each group is specialized in a precise topic, including chemical recycling of PET.

4.1.4 CEFLEX (Europe, 2017)

CEFLEX is a nonprofit consortium of packaging suppliers, manufacturers and brands, which decided to focus on the recycling and improvement of flexible plastic packaging. Their objective is to include 80% of the flexible packaging in a closed loop economy.

Only a few NPO acting for packaging sector improvement were presented in this part. For instance, WRAP is another NPO, based in UK, which participated in the creation of the UK Plastic Pact, in collaboration with the Ellen MacArthur Foundation. The Association of Plastic Recyclers provides guidelines for plastic recycling use and supports its development worldwide. In the same vein, Plastics Recyclers Europe, based in Belgium, launched recently its "Recyclass" platform, with the objective to push companies to create and use plastic packaging respecting recycling criteria in Europe. All these organizations have the same objective of improving the plastic packaging sector, thanks to guidelines and other actions and projects. Their work allows companies to implant precise targets concerning the materials and end-of-use of their packaging. However, differences exist in the way of acting between these associations. In 2020, the European Plastics Converters association decided not to sign the European Plastic Pact proposed by the Ellen MacArthur Foundation. According to the association, the objectives are unfeasible for this short deadline, and would not be helpful for the sector in the long run [106]. For the European Plastics Converters association, Europe should focus on plastics landfilling problem and bad wastes management first.

4.2 Brand companies

This part refers to companies which sell products to consumers and succeeded in creating a brand.

4.2.1 Products improvement

Since several years, environmental impact of plastic packaging, on resources, pollution but also marine litter, started to emerge in the developing path of brand companies. They started to change materials and reduce impacts of their goods, without modifying the quality and the user experience. To this end, brand companies decided to work in collaboration with NPO, to develop feasible and ecological objectives regarding plastic packages, but also other environmental topics. In order to be as transparent as possible, many of these brand companies share their sustainable report annually, containing their progress and objectives on environmental points. These sustainable reports are open to the public and available on brands website. These are good sources of information about brand implication in plastic packaging circular economy development. For instance, the Coca-Cola Company aims to make 100% of its packaging recyclable by 2025, and use 50% or more of recycled content in packaging by 2030 [107]. Cosmetic brand L'Oréal objective is to make, by 2025, 100% of its plastic packaging recyclable, refillable, compostable or reusable [108]. Some companies created a list of materials and products to eliminate before a certain deadline. This is the case of Nestlé, which decided to eliminate all its products made with PS and PVC before 2022 [109]. On this principle, many brand companies decided to make and communicate their commitments regarding plastic packaging. However, these reports, and objectives in general, have to be carefully interpreted. When a brand announces 100% of recyclable products, it does not mean that 100% of its products will effectively be recycled. The effective recycled percentage depends on the location, the contamination and the sorting. Theoretically, a LDPE film is recyclable, but practically, because of contamination, leakage in environment and bad sorting, this plastic film has a low probability to be recycled. This is also the role of these brands to integrate the end-of-use of their products in their organization, which includes consumers orientation.

4.2.2 Influence on consumers

How2Recycle (US), Info-tri (France) or On-Pack Recycling Label (UK), recycling labels are nowadays present in many countries. By applying these labels on their products, brand companies help consumers to make the good choice for wastes sorting, and avoid contamination of recycling streams.

Besides products end-of-use, companies can also influence the way of consuming among the population. It can be done by modifying the materials used, but also shape and dimensions of plastic packaging. This modification already started in several sectors, from soap refills, requiring less plastic and only one material for the pouch, bigger volumes for beverages and liquids in general, or plastic replacement by cardboard or paper alternatives. The refill alternatives require plastic pouches, which can be invalid for recycling. But this option reduces noticeably the consumption of plastic compared to a classic package [110].

More and more brand companies start or continue to develop the ecological aspect of their plastic packages. This behavior falls within a current green tendency, which could have a positive impact on the plastic packaging sector. However, these packaging modifications operated by brand companies have to keep environmental improvement as unique target. Hence, each decision regarding this topic has to be thoroughly evaluated, not only for an immediate effect and to satisfy consumers, but also for the future. Greenwashing is a term designating an ecological policy, whose only purpose is to sell and depict a company as a green one, and not to have a positive impact on the environment. This greenwashing practice has to be averted in order to avoid a displacement of the existing packaging problem to another sector.

4.3 Packaging materials suppliers and manufacturers

This category regroups producers of packaging materials, which collaborate with brand companies to supply packaging for their products. DIC Corporation is a packaging, coating and inks supplier for numerous brands, that convert its products in packaging solutions. The role of DIC Corporation and other materials suppliers is important in the general process, and they are inseparable from the sustainable transition. Indeed, brand companies place their materials on the market, which end in bins and sometimes in the environment. In order to limit the global impact, suppliers have to find greener alternatives to the current materials, and allow brand companies to propose products with more sustainable packaging to customers. For this purpose, suppliers started to focus on new materials or modify existing ones to limit their impact. In this respect, paper alternatives were introduced. Paper does not consume oil as raw materials, has a faster decomposition rate which limits the impact of environment leakage and is highly recyclable. Indeed, paper packaging and containers recovery rates are 73% in the US (2017, 13% for plastic) and 81% in Japan (2017, 22% for plastic) [79][80][111]. The case of paper packaging will be described more in details below, in 5.2.2.

Packaging material suppliers focus also on the use of recycled content for plastic packaging. As described above, their use for direct contact with food and other delicate products is still limited. Indeed, PET is nearly the only one that can achieve these applications. But for other applications, suppliers are developing and increasing the use of post-consumer recycled (PCR) content. Recently, UPM and Sabic launched a packaging label made with 100% of PCR polypropylene [112]. The recycled and virgin materials are nearly alike in properties. But PCR plastics development is still in progress, in order to be used in more packaging types and increase the material quality. On top of that, PCR content price is another problematic. Even if the sustainable and ecological argument is important, companies also act depending on the cost of a material. PCR content is often of lower quality than its virgin equivalent, and price is then an important argument in the decision of choosing one or another material. In 2018, recycled PET price was around 1,200 $\$.ton^{-1}$ and virgin PET around 1,400 $\$.ton^{-1}$ [113]. But, in 2019, prices for recovered PET outreached virgin PET ones [114]. This could have influenced small companies in their choice to switch from PCR PET to virgin PET. Because larger companies often keep a fixed budget for their raw materials, including PET, this decrease of virgin plastic price allowed these companies to allocate a bigger part of the budget to recycled raw materials. Indeed, recycled PET price stayed stable in 2019. It had a good impact on the recycling market. In March 2020, the prices for recycled and virgin PET flakes were respectively around 980 $\$.ton^{-1}$ and 870 $\$.ton^{-1}$ for virgin one [115]. The prices difference is higher and it could have an impact on the recycling market.

Indeed, the COVID-19 crisis overthrew oil prices, and therefore virgin plastic prices. If companies stay focus on their sustainable objective, this price difference should not have an impact on the recycling market. COVID-19 epidemic also had an impact on the material quantities. At the present time, the recycling market is lacking of material. This is due to several phenomena, including stockpile and sorting stop, conducting to a decrease of wastes entering the recycling path. Effects of COVID-19 epidemic on recycling market are not yet known, but a decrease in collected and sorted plastic wastes could lead to an increase of the recovered plastic price, and a decrease of the recovered materials production. Even if companies with a fixed budget, as seen above, would certainly continue to use recycled plastics, even with higher prices, the reduction of available recovered materials could push them to use virgin plastics. Nowadays, recycled content in packaging can be used as a selling argument to increase packaging prices, and participate to the development of the recycling sector. Thus, even with the current crisis, it is important for companies to invest and support the plastic recycling sector, in order to allow packaging suppliers to develop the use of recycled content.

4.4 Governments

Governments, through laws and taxes, can influence companies selling packaging in the territory, but also population and its way of consuming and sorting wastes. These entities have maybe the largest potential impact compared to other entities of this section.

4.4.1 Extended producer responsibility

Extended producer responsibility (EPR) is a concept introduced in Europe in the 1990's. As its names indicates, it extends the responsibility of companies which introduce products, that will become wastes after being used by consumers [116]. EPR is paid by companies to governments, and allows to cover part of the wastes management costs. The amount paid by each companies depends on the quantity of wastes produced, but also its nature. Thus, the more sustainable is the packaging, and so the lower its impact on the environment, the lower are the EPR fees. EPR objectives regarding packaging wastes are clear [117]:

- Cover wastes management costs.
- Push companies to create sustainable packages, with a lower environmental impact.
- Increase recycling rates and develop recovery of these wastes.

Besides taxes, EPR can take the shape of deposit systems. In this case, companies have to implant infrastructures dedicated to the collection of targeted plastic packages. The EPR form depends on each country. For instance, in France, for households' packaging, producers are only responsible on a financial aspect, whereas in Germany, in addition of the financial responsibility, producers are also physically responsible [118]. Thus, they have to implant collection system for their packages. This responsibility can become complicate for producers, which can decide to manage EPR alone or in group. In this last case, the group of companies often act under the management of a superior entity, which collects and organizes the different actions. This is the case in France with the CITEO group. EPR participation is often indicated with a logo, directly on packaging. There is an ambiguity with the Green Dot logo used in Europe, which must not be confused with a logo indicating that the packaging has to be recycled or is recyclable.

Figure 30: Green Dot logo used in Europe to indicate EPR participation

Indeed, according to a survey conducted by Amcor company, only 15% of the European interviewees knew that this Green Dot symbol indicated EPR, and not the recyclable characteristic of packaging [119]. The concept of EPR is born in Europe, but countries worldwide created laws based on this concept. In Japan, the *Container and Packaging Recycling Law* was implanted at the end of the 1990's. Producers of packaging wastes have to pay a tax to allow and develop the recycling of their products [120].

Despite the bad aspects of EPR, especially its heterogeneity regarding implantation, impact and costs management, this measure helps to create a better waste management system. It pushes companies to develop more sustainable packaging solutions, with ecological design.

4.4.2 Wastes management development

Even if governments do not manage directly the recycling of packaging wastes treatment factories, they can implant measures in order to develop the sector. It takes place through collection improvement, population awareness, but also stricter rules.

One of these measures is landfills ban. As depicted in the previous section, landfills are a loss on several aspects. This management method does not produce resources from packaging wastes, neither material nor energy, and it requires a lot of space. Because wastes, especially plastic wastes, degrade slowly, they pile up in landfills, whose size has to be increased year after year, and closed when full and that further extension is not possible, in order to open a new landfill. This management, though an ecological disaster, can be viable in the long run for countries with sufficient exploitable area. This is one of the reasons why landfills are so popular in the US (Figure 31), with around 70% of the plastic packaging wastes sent to landfill [79].

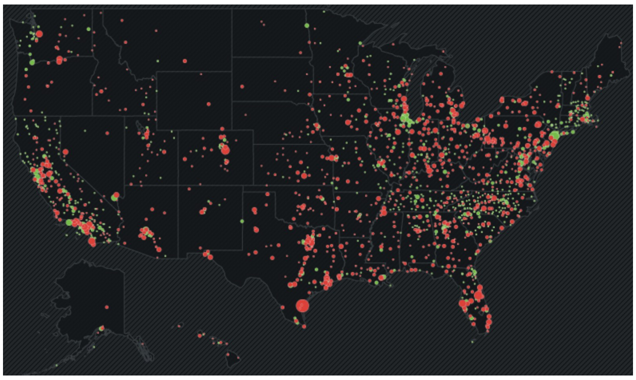


Figure 31: Distribution of closed (green) and open (red) landfills in the US [121]

However, all the countries in the world are not in the same situation than the US, with nearly half of the country area being vacant. Driven by this reason, among more ecological ones, several countries started to ban landfills on their territory. In fact, these countries banned the disposal of common wastes (understand non-hazardous wastes, that can be either burnt or recycled) in landfills. Some wastes types, including ones generated by EfW plants (cf. 3.4.2), cannot be burnt and still have to be disposed in landfill.

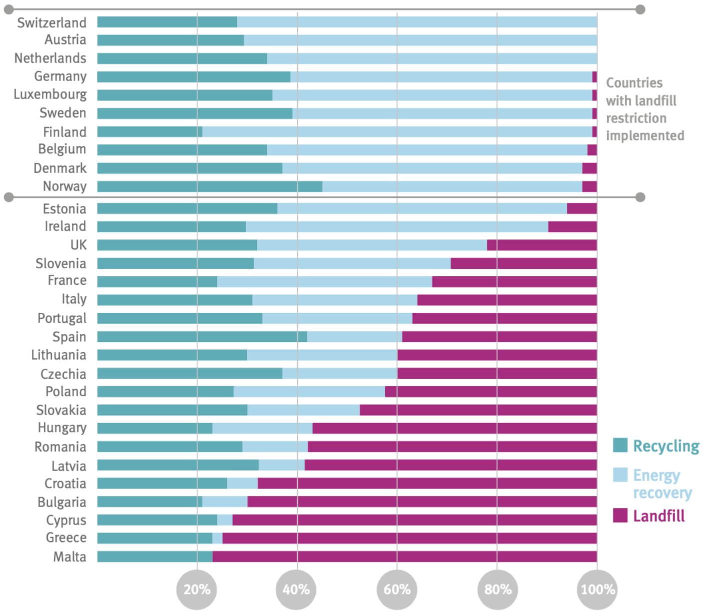


Figure 32: Disposal of post-consumer plastic wastes in Europe (2018) [122]

However, by banishing disposal of households' and common wastes into landfills, these countries have to develop recovering methods, which leads to a better plastic packaging recycling compared to other countries [122]. Figure 32 shows that several countries of Europe have implanted a ban or limitation regarding landfills. In Europe, target was fixed to stop the recyclable wastes disposal in landfills by 2025. The European commission is running this decision that should be a huge ecological improvement. Indeed, by banishing recyclable wastes sent to landfills, Europe could largely increase its production of recovered materials and energy from wastes burning [123]. Thus, the amount of plastic wastes sent to landfills largely decreased in Europe during the last few years. This reduction shows that the objective of zero recyclable wastes in landfill by 2025 is achievable. Several countries already decided to ban recyclable wastes disposal in landfills. In Switzerland, since 2000, it is forbidden to send burnable wastes, that can be used to produce energy, into landfills. In Austria, only a small percentage of wastes are sent to landfills, and these plants are subjected to an important tax. Austria uses this tax revenue exclusively to decontaminate landfills.

With EfW and material recycling plants development, landfills should progressively decrease worldwide, in order to reduce the amount of stowed wastes and produce resources, materials or energy.

4.4.3 Wastes exchanges and China's ban

From shelf to bin, the packaging journey is often simple and quick. However, its journey from bin to end-treatment is generally more complicate. Indeed, plastic packaging wastes generated in a country are not always treated on the spot. Worldwide, countries export and import plastic wastes. Exporters want to shed their troublesome

and numerous wastes, whereas importers want to use them as material source, to produce recovered plastic that they can sell for a higher price. In early 2018, in the European Union, the price for one ton of plastic wastes was around 300€ [124]. The two main exporters of plastic wastes in the world are Japan and the US.
Since the 1990's, China was the main importer of these wastes, with nearly 50% of all the post-consumer plastic wastes produced from this period to 2018 [125]. However, Chinese government did not want its country to accept the waste of developed countries any longer. Thus, started from the 1st of January 2018, China decided to reduce drastically its plastic wastes imports. Indeed, on this date, China started to ban plastic wastes imports. This banishment touched all the plastic wastes, besides ones coming from the industrial sector. As explained above in this report, because of low contamination, larger size and homogeneity of these plastic wastes, their recycling is less problematic than for post-consumers plastic wastes. As China was the first importer, this ban had a consequent impact on other plastic wastes importers, mainly present in Asia. As depicted in Figure 33, even if China's ban engendered a decrease of plastic exports worldwide, it was accompanied at first by an increase of the imports in other countries. Malaysia, Thailand and Taiwan are particularly touched by the Chinese measure [126]. In these countries, the weight of plastic wastes imports has at least doubled, and the management of this increased quantity creates numerous problems for these countries.

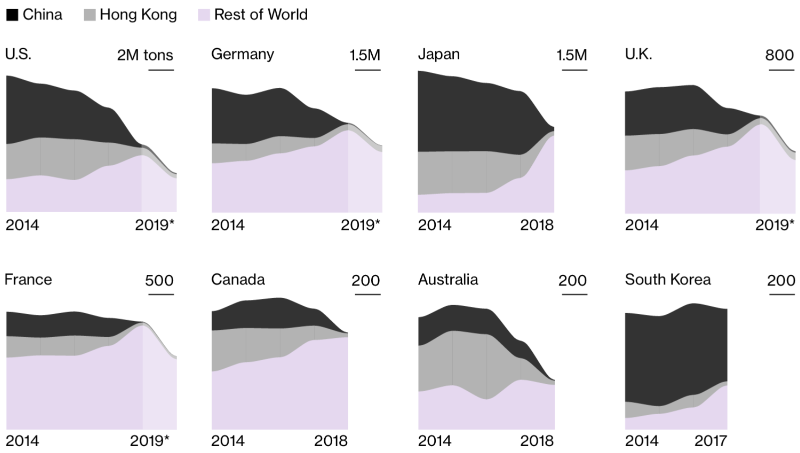


Figure 33: Change in plastic wastes exports after China's ban [127]

Even if there was a reduction in the exported amount, the quantity is still too important to manage for these countries. By banning plastic wastes imports, China broke a stable process which was implanted many years ago. The decision was announced by the Chinese government before taking effect, but the time was too short for exporters to rethink completely their wastes process. Thus, they switched to the easier solution: developing countries that still wanted to buy their plastic wastes. The switch being to prompt and enormous for these countries, it led to mismanagement and complications in plastic wastes treatment. These complications are still present. Several factories in Malaysia, which are processing plastic wastes imports, accumulate plastic wastes that will be treated several months after their arrival. In this context, Malaysia and other Asian importers decided to act. They sent back plastic wastes containers to exporters when they did not respect the export criteria. Because Chinese decision happened quickly, storage problems occurred in exporting countries, leading to bad sorting. Malaysia being already overwhelmed by the quantity, they cannot manage illegal plastic wastes imports, for instance mixed with paper wastes. The country already sent back 150 containers of these wastes, representing nearly 4,000 tons. Among these containers, more than 80 were sent back to UK and France [128].

In the end, China's ban is certainly a positive decision for worldwide plastic waste management. This decision pushes exporting countries to reduce and manage their plastic wastes inside their country, one possible impact being an increase of recycling facilities and process efficiency. The same improvement could be observed in importing countries, including Malaysia. However, Chinese decision took effect too promptly. A progressive ban would have given time to both exporters and new big importers to develop infrastructures and processes. Moreover, a decrease of recycled content is expected, which could conduct to an increase in prices [129].

4.4.4 Single-use plastics ban

As depicted in the introduction, one of the main concerns regarding plastic nowadays is its bad management. Especially, the effect is dramatic when these wastes leak in the environment, including oceans. However, all plastic wastes types are not equal regarding this mismanagement. Most of plastic packaging belongs to the single-use plastic category, with straws, cups, cutlery and bags. Because of their small size, low weight and the fact that they can be used during transport, this category has a higher risk to leak into the environment. Hence, several governments started to act in order to reduce the impact of SU plastics.

It started few years ago, by single-use plastic bags banishment. The first country was Bangladesh, in 2002, which decided to ban these bags, allowing only thicker ones, supposed to be used several times [130]. Since then, different policies were implanted in the world to treat the plastic bags problem. In the US, there is no national ban on SU plastic bags, but local legislations exist. They covered around 10% of the whole population, and mainly imply SU plastic bags ban and fees for sustainable or reusable alternatives [131]. In Japan, there is actually no ban on these bags. Businesses alone decide and some brands already stopped to provide plastic bags, replacing them by paper alternatives. In supermarkets, symbolic discount are often applied when customers decide to avoid plastic bags. However, the amount is paltry, around 2¥, representing less than 0.02$. Since July 2020, retailers have to charge customers for these SU plastic bags, minimum amount imposed by Japanese government being 1¥[132]. But this charge system concerns neither biodegradable plastics, including bags made with PLA, nor businesses other than retailers. In Europe, several countries have banned SU plastic bags years ago. For instance, France voted this ban in 2014, which took effect in 2015 [133]. Same law was implanted in Italy at the beginning of 2010's.

SU plastic bags ban was the first measure taken by countries in the aim of banishing all SU plastics. Countries want to reduce their use of SU plastic and wastes generation. Thus, bans on SU plastics are in development worldwide. In France, since the 1st of January 2020, cotton buds, plates and cups made with plastic are banned [134]. More SU plastics, including straws, will follow in the next years. France anticipates European law, voted in 2019, which bans single-use plastics in European Union by 2021 [135].

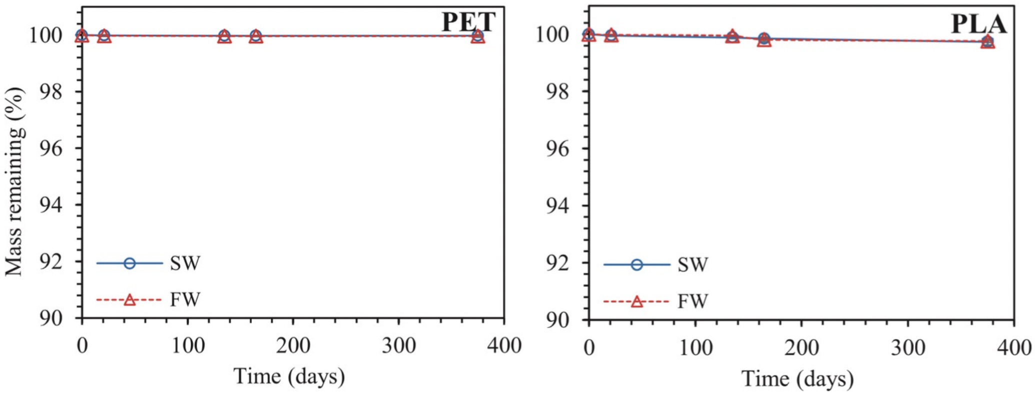


Figure 34: Degradation of PET and PLA in fresh (blue) and sea (red) water, at 25°C [136]

However, even if these bans are an achievement in tackling the plastic marine litter and plastic pollution in environment in general, there is still a long way to go. As seen above, these bans barely concern biodegradable plastics. Admittedly, these plastic materials have a shorter decomposition time than common plastics, but it requires special conditions, rarely reached in natural environment [136]. This is an important notion to remind, in order to stay focus on the real potential of these biodegradable alternatives, which is to produce less waste by an improved degradation process in compost facilities. Experiences were conducted in fresh and sea water in order to observe the degradation time of different biodegradable polymers. As seen in the previous section and Figure 20, PLA composting at T = 58°C and increased humidity leads to an important degradation of the material in 30 days. Even if microplastics are still present after this period, the plastic packaging has achieved a correct degradation. Figure 34 depicts the degradation of both PET and PLA films in water at 25°C. Figure 35 describes the same experiment in the case of polylactic-co-glycolic acid (PLGA), a copolymer of PLA. It is clear that the fast degradation phenomenon observed in compost factories is not observed in this case. Moreover, products under experiments here are films, whose thickness is lower than 1 mm. Because thickness (and mass in general) has an impact on biodegradation, it is accurate to predict that results highlighted in Figures 34 and 35 would be longer with bottle package as presented in Figure 20. However, the results are already significant. After one year, PLA has nearly not been degraded and it took 270 days for PLGA in both fresh and sea water to completely degrade.

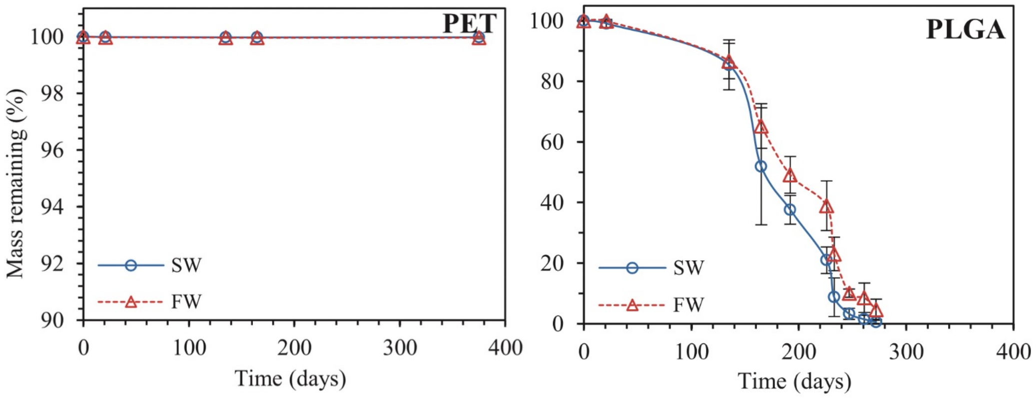


Figure 35: Degradation of PET and PLGA in fresh (blue) and sea (red) water, at 25°C [136]

Results in natural environment would certainly be different, especially in oceans due to natural flow agitation and UV exposition that could help for biodegradable polymers decomposition, but it would not engender a drastic change in the required time.
Another difficulty with bans implantation is legislation itself. In the US, laws exist that banish bans [137]. Companies with interest in keeping the use of plastic bags utilized this law to break ban in a city in Texas state. Same event happened in cities in Colorado state, despite the ecological benefits of this kind of measures. It is certain that government has to act in this situation, to prevent companies economically implied in this kind of decisions to have damaging impacts in the sustainable evolution of cities or even states.

All the entities presented in this section have a major role to play in the plastic packaging sector evolution. They are all linked and have, in different ways, an influence on population behavior, which means plastic packaging consumers behavior.

5 Possible improvements of the global packaging situation

At the present time, technologies are developed or improved in order to facilitate the end-treatment of plastic packaging wastes. These technologies can occur at the beginning of the plastic packaging chain, with biodegradable materials or greener alternatives, or at the end, with recycling or thermal recovering. They allow to reduce, change the nature or create added value from these wastes, but not to recover them completely. The current plastic packaging sector is more likely to reach an open-loop recycling system than a closed one.

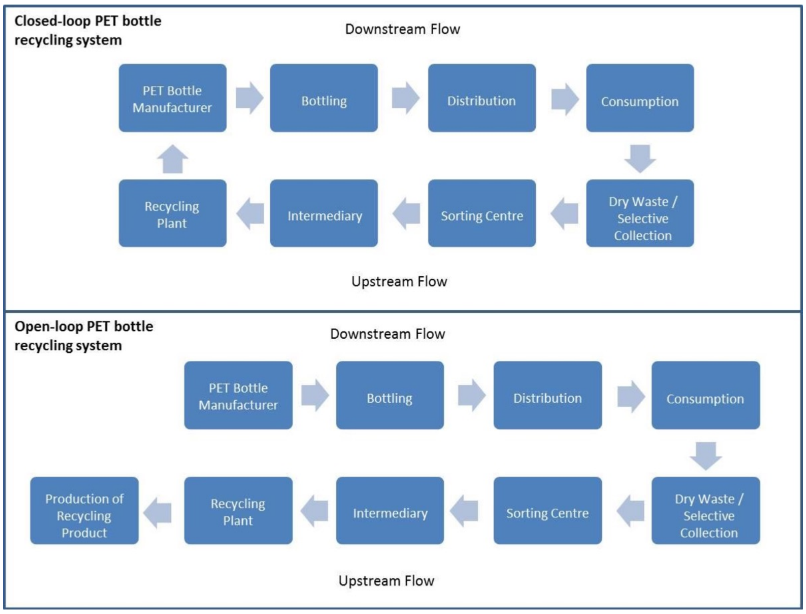


Figure 36: Open and closed loop recycling for PET bottle [138]

The concept of open-loop recycling supposes that the plastic material undoubtedly ends in a facility that does not produce the same new material from it, mechanical recycling or thermal recovery in the best case, landfill in the worse, because quality of the recovered polymer would not be sufficient to create a new plastic product. At the present time, nearly none of the plastic packaging materials are included in a closed loop recycling process, and only a small portion, as seen above, belongs to the open-recycling system. In fact, a true closed-loop process, where one material could be indefinitely recovered into a material of same quality, is utopian. Recently, only PET bottles are approaching a closed-loop system. However, even this packaging type can be recycled only few times into a new bottle, before leaving the loop because of poor quality. In order to connect to this closed system, several point in the existing sector need to be changed. The existence of several challenging products is in total opposition with this closed-loop concept, and there is a dire need to provide materials and recycling innovations to deal with them. Challenging packaging and potential innovations will be presented in the next parts of this section.

5.1 Challenging plastic packaging products

Different types of plastic packaging are the most problematic and challenging in terms of recycling. The reasons can be the nature of the material used to create the product, the assembly of the product, or even its use, which engenders complications for the end-of-use treatment. These packaging products are part of the *redesign* category presented in the introduction of this paper. They cannot be reused, but they require a deep improvement of their nature, in order to improve their end-of-use path and reduce the plastic ecological problem. These packaging products represent at least 50% of the packaging items made with plastic, and more than 30% of the market by weight [139]. For this reason, their redesign is one of the main priorities in the challenging plastic situation.

5.1.1 Multi-material

Multi-material packaging products account for around 13% of the global market by weight [139]. These materials consist in combining layers or parts of different types of plastics, or other materials, in a single packaging (Figure 37). The purpose is to increase or create new properties. It is an upgrading process of mono-material properties. Thanks to this process, new properties can be achieved, including shape retention or water and moisture barriers. Multi-material packages can get up to nine consecutive layers of different materials.

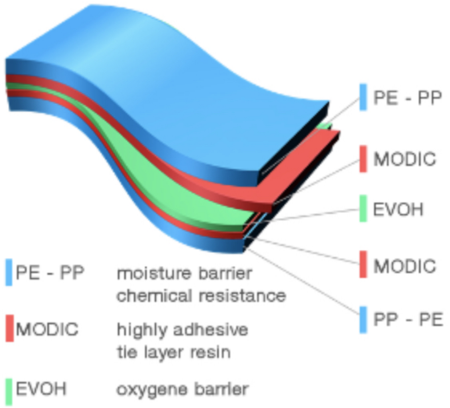


Figure 37: Multi-material packaging layout [140]

However, by combining materials of different nature, including metal-plastic or paper-plastic combinations, the recycling process becomes drastically more complicate and, in some cases, impossible. Of course, these different materials cannot be recycled in the same stream, even if a small contamination percentage is accepted. To achieve their recycling, separation of the materials is required. It intensely increases the difficulties encountered for material recycling of these packages, which is not cost-effective for companies [141], that conducts in most cases to incineration or thermal recovery. They are commonly present in the food sector, including aluminium and plastic combination for food pouches.

These multi-material packages are really appreciated by manufacturers, because they allow to create thinner packaging than mono-material ones, which conducts to a reduction of the material resources demand and greenhouse gases during the transportation due to their light-weight nature. These products are composed of three up to nine layers of different materials, with resins across these layers. There is no official and standard composition, each manufacturer can create the multi-material according to its wishes, which complicates even more the recycling process, especially to identify resin composition. Moreover, because they are frequently used for food pouches or light-weight food products, it engenders complications for collection and sorting. Because they belong to the food packaging category, numerous users are not aware of the recycling process and in which bin throw these packages. They are also problematical during the sorting step. Indeed, due to their light weight and 2D format, they are often considered as paper by sorting captors, and can contaminate this other recycling stream.

5.1.2 Small format

This category represents around 10% of the weight of the plastic packaging market [139]. Belong to this segment lids, straws or wrap films. The main problem with this small format packages is not their nature, because often made with mono-material plastic (PP or PE), but their size, that makes them easily escape from the waste stream and leak into the environment. These small format single-use plastic products are among the first sources of plastic marine littering. Even if technologies allow to recycle these products, they are seldom recycled. Indeed, during the sorting process, one of the first step is the sorting of small contaminants thanks to a mesh. The mesh used have a standard size of 4 to 7 cm. Because of their size, small-format packages remain trapped in this mesh, like stones or other small contaminants. But it is not worthwhile for sorting companies to add another process to sort these little plastic items from contaminants in order to be recycle. It is not profitable due to the low quantity and value of small format packaging. In most cases, these packages end up in EfW factories or even landfills, where they are susceptible to leak in nature.

5.1.3 Uncommon

Uncommon plastic packages account for 10% of this global market by weight [139]. The three main plastics of this category are EPS, PVC and PS. They represent around 85% of this uncommon packaging sector. In the first section of this report, reasons were highlighted for the poor recycling rate of these polymers. As for small format products, their volume is too low to allow their recycling to become worthwhile. Indeed, they are often used as consumer products. But when used as business products, their volume is higher and they are collected in a dedicated stream, not mix with other plastics. In this case, it becomes interesting to recycle these unusual products. Moreover, these materials are frequently responsible of other streams contamination. Thus, they can reduce significantly the quality of other recycled content. There is also a concern due to the hazardous behavior of several uncommon plastics, including PVC, which contains carcinogenic monomers. These unusual plastic packages are usually sent to EfW factories or landfills.

5.1.4 Contaminated

The proportion of these packages is difficult to quantify, but this category contains all the plastic packaging contaminated with organic content during use or after. The recycling of these products remains complicate because the organic elements attached can be impossible to separate. Organic elements could lead to contamination and reduction of recycled content quality. Thus, these packages also end up burnt or landfilled.

5.1.5 Black plastic

This is one of the main problems encountered by sorting factory in the plastic recycling circuit. Currently, these factories often use a Near Infrared (NIR) sorting process (cf. 3.1.2). But at the moment, only a small portion of black plastic packages can be sorted thanks to this process. Indeed, most of the black plastic products are made with carbon black pigment to obtain this black colour. Manufacturers choose this pigment because it is cheap, allows food contact and has a high coloration strength [142]. The problem with this carbon black pigment is that, as for other common black pigments, it has a high absorption in the infrared range. When going in the NIR sorting chain, it is not detected, and cannot be recycled. Each year in United-Kingdom, this sorting problem concerns in between 30 and 60 kt of plastic wastes.

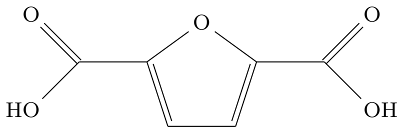
5.2 Materials

5.2.1 Plastic material improvements

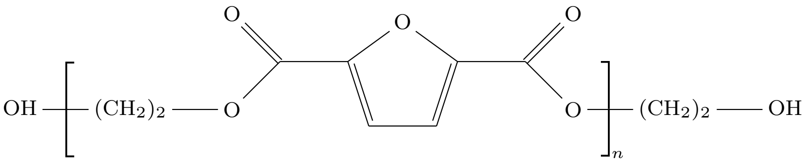
Regarding packaging of the precedent part, there are several ways to improve the nature of these products by changing materials. Black color for packaging is a selling and marketing argument for brands, and despite its current controversy, these companies still want to use it. Thus, packaging suppliers started to develop black pigments that can be sorted by NIR sorting systems, while providing the same result in terms of color than black carbon pigment. Recently, Colors & Effects company, belonging to BASF, announced launch of a new pigment, suitable for common packaging plastics and, more importantly, that can be detected by NIR sorting [143].

Problematic linked to multi-material is more complex. Unlike black pigment, the use of different combined materials allows to increase barrier properties, to reach ones that could not be possible with only one material of the same size and thickness. These properties are essential for protection of the product, and not a selling argument. To solve this problem, it is interesting to come back to mono-material plastic packaging. By selecting the better polymer type for a dedicated application, it is possible to create flexible packaging made with a unique material. Coating is often necessary to improve properties, but the amount is low compared to main polymer used, and the impact in recycling process stays reasonable. It also possible to use coating made with the same material than the main part of the package. Recently, Borealis launched a new pouches packaging collection, with pouches made with mono-materials PE and PP to improve recycling [144]. This company also removed the aluminum layer often present in pouches, which is inseparable from plastic material in common recycling chains. However, mono-material packaging are still made with several layers, but with the same polymer. The main problem encountered recently by manufacturers, with mono-material packaging production, is the sealing behavior. The most common process for sealing is thermal sealing. It allows to provide closure to packaging and protect the content. In multi-material packages, a special polymer layer, with high heat resistance, is used for sealing. Polymers used in mono-material packages have a lower heat resistance than sealable materials contained in multi-materials packaging. The difficulty is to provide good sealing at large scale, in production chains, without degrading the quality of the final product due to too high temperatures or poor sealing [145].

Another option should be to use bio-based plastics. The notion of bio-based plastic was introduced above with PLA (cf. 3.3.2). In the case of PLA, the interest for composting was not really relevant, because of poor sorting methods and precise conditions required. However, bio-based alternatives to petrobased plastics could be a good opportunity to increase the sustainable aspect of the packaging sector. This change in the material would have to be developed at large scale without implying a decrease of the properties. Since several years, the development of a certain bio-based polymer to replace PET is studied. This bio-based polymer is called polyethylene furandicarboxylate (PEF). PEF is bio-based but not biodegradable, compared to PLA. It belongs to the polyesters family and its chemical structure is really closed to PET one, the main difference being that the phenyl group is replaced by a furan group.



Furandicarboxylic acid to replace terephthalic acid



Polyethylene furandicarboxylate (PEF) to replace PET

PEF is obtained from furandicarboxylic acid and ethylene glycol, through the same reaction mechanism than for PET formation. Furandicarboxylic acid is bio-based and produced from fructose, that can be obtained from natural sources. This is not the case of terephthalic acid, used for PET, which is petrobased. Moreover, PEF properties are better than PET ones, including barrier properties, which are key properties for bottles, the main packaging application of PET. Indeed, in the case of PEF, compared to PET, CO2 permeation is decreased by 56% and O2 permeation by more than 89% [146]. It also has a higher T_g, a lower T_m and a higher Young's modulus [147]. Regarding the ecological benefits of PET replacement by PEF, LCA studies were realized to calculate its impact. By comparing energy requirements and green house gases emissions, PEF seems to have a lower impact than PET. Indeed, both required energy and emissions are halved in the case of PEF [148]. The recycling process of PEF is also similar to PET one, and PET recycling stream contamination by PEF is possible and acceptable if limited at 5% [146]. Hence, PEF could be a good replacement material for PET, by reducing ecological impact while getting better properties. However, there is currently no dedicated collection, sorting and recycling path for PEF, and one must be created to avoid PEF-based products to end up in EfW plants or landfills as PLA. Moreover, the LCA analysis for the ecological impact of PEF does not take into account the effect of fructose production (for furandicarboxylic acid production) on land use, including deforestation.

NIR detectable black pigment and bio-based or mono-material plastic packaging could play soon an important role in the improvement of this sector. However, they cannot solve problems linked with neither small-format nor contamination. Increasing packaging size with only purpose to make it more detectable, and avoid leakage in environment or escape from sorting technologies, is a nonsense. Concerning organic contamination, this problem is linked to the natural usage of these packages. As long as collection processes and recycling technologies cannot sort and recycle these problematic plastic packaging products, and avoid their leakage into environment as litter, the solution could be to change plastic for more sustainable alternatives.

5.2.2 Paper alternatives

A new trend emerged in the packaging sector several years ago. This trend is the use of paper as main or mixed material, in replacement of plastic. Paper and cardboard are already used as packaging for protection, transport, beverage cartons and sometimes food (flour, pasta and dry food in general). In 1999, in Europe, it represented 40% of the total amount of materials used for this sector [9]. Worldwide, in 2016, around 36% of packages were made with these wood-based materials [149].

The most recent and drastic change from plastic to paper occurred with shopping bags, but this phenomenon starts to reach more and more packaging types. These pulp alternatives enjoy a good popularity among customers, which participated in the shift of many companies from plastic to paper packages development, as a selling argument. This shift allows to reduce the amount of plastic created, and that can eventually leak into the environment. La Roche-Posay, L'Oréal cosmetic brand, recently launched a sunscreen tube partly made with cardboard, which allows a 45% reduction of plastic use for this packaging. Moreover, recycling rate of paper and board packaging is high (Figure 38). This rate is largely higher than plastic

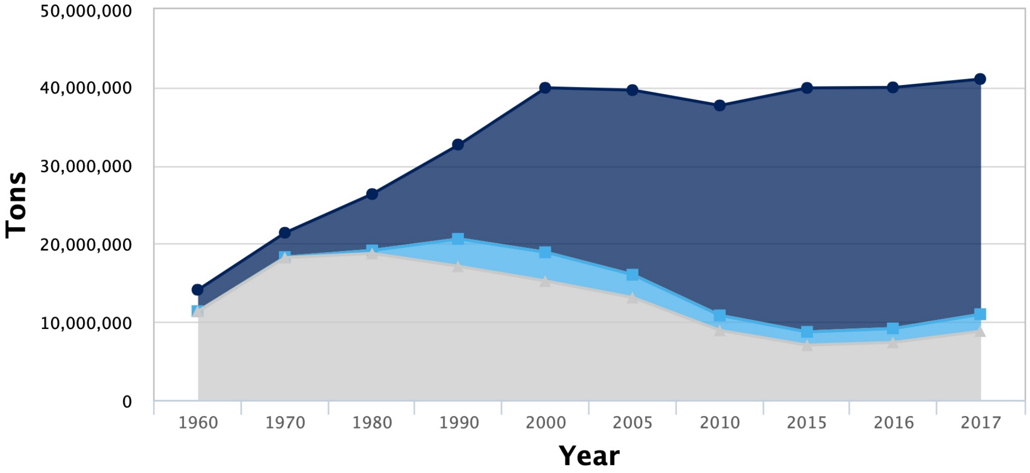


Figure 38: Recycling (dark blue), EfW (light blue) and landfilling (grey) of paper and board packaging wastes in the US [79]

packaging one. However, this alternative is not free of controversy, and it is not as easy to replace every plastic package by paper one.

The first problem with the use of paper as replacement, is the fact that it requires plastic coating to provide properties in the same range than plastic. It is not possible to use raw paper or board for packages needing gas or vapour barrier, and mechanical properties are also lower compared to plastic. For instance, paper has a MVTR of 690 $g.m^{-2}$ [150], whereas for synthetic polymers studied above, despite PS known for its low MVTR value, all have a MVTR around or below 100 $g.m^{-2}$. Spaces in paper molecular structure let gas molecules pass more easily, and the hydrophilic groups that composed paper (cf. 5.2.3.1) are responsible of its weak water vapour resistance. In order to increase these properties, paper is commonly coated with polyolefins, but it inhibits recycling. Indeed, even if processes for paper body and plastic coating separation exist [151], they are not frequent, and most of municipalities and sorting companies do not accept polyolefin coated paper for recycling. Indeed, general guidelines regarding paper recycling do not consider coated paper as recyclable [152][153]. Currently, an important part of the plastic coated paper wastes are sent to EfW or landfills, despite high recycling rates of raw paper and board packaging highlighted above. When there is a commercial interest to separate paper and coatings, including beverage cartons with aluminium layer, the whole product is shredded, and the different materials can be separated by flotation. Moreover, despite difficulties for recycling, polyolefin coating reduces the sustainable aspect of paper alternatives. Even if studies found that coating does not prevent paper natural degradation [154], coating is still present at the end. Thus, if coated paper package leaks in the environment, the littering impact will be lower than for plastic package, but still existing. Indeed, plastic coating can represent around 5% of the total package weight for cardboard with one coating layer, and more than 20% for beverage cartons [155]. This percentage is not negligible.

The second problem with paper alternative is that consequences of its manufacturing on the environment are not clearly known. Comparison between a PE bag and its paper alternative, containing 30% of recycled paper content, shows that the resources consumption is higher for the paper bag than for plastic one (Table 9), including fossil fuel use [156]. Energy consumption includes production and transport. These results were obtained by Life Cycle Analysis (LCA). LCA is a useful tool to compare environmental impact of products.

	Energy (MJ)	Water (L)	Fossil fuel (g)	Waste production (g)
Paper bag	2.6	3.8	23	34
PE bag	0.7	0.2	15	7

Table 9: Necessary resources for production of one bag (with same carrying capacity) [156]

Even if the results depend mainly on the databases used and that they do not prove clearly that paper bags are worst than plastic ones, they have to be taken into account. Worldwide plastic packaging consumption reaches astronomical quantities, but its effects on environment are quite well known. It is not the case of paper packaging sector. Replacement of more than 100 million tons of plastic packaging produced annually by paper alternatives could surely divert million tons of plastic leaking into the ocean as marine litter, but it would certainly imply another unknown environmental cost. These packages, as plastic ones, are currently not recyclable, and their end-treatments are mostly EfW and landfilling. Thus, increasing paper products production will require to cut more trees. At the present time, several billion of trees are cut each year for paper packaging production. This replacement would also process more wastes, knowing that more material is required for paper packages compared to plastic ones, to achieve the same properties. Moreover, these wastes have a lower heat value than plastic for thermal recovering.

In this context, paper itself is not the perfect candidate for plastic packaging replacement. Indeed, it uses plastic coating to achieve acceptable properties, and its positive effect on the environment is not clearly highlighted because of poor, next to nil, recycling. To improve the situation, suppliers should use a material as sustainable as paper, in case of leakage into environment, but with properties similar to plastics and a good recyclability.

5.2.3 Nanocellulose opportunity

5.2.3.1 Material presentation

Nanocellulose could be a good option concerning properties close to plastic ones and good recyclability, while combining a more sustainable impact. The term *nanocellulose* can describe different types of cellulose, under fibrils or crystal form. In this report, it will refer to cellulose nanofibrils (CNF). Several other names can be used to describe nano-size cellulose fibers, but this report will only use CNF denomination. CNF come from cellulose, which is produced by different sources and found in natural environment. The main source of cellulose in the industrial sector is wood. Cellulose fibers are constituents of wood and CNF are nanoscale fibers obtained from cellulose thanks to different processes [157]. The type of process used for CNF production depends on the wanted final properties of the material. Some methods have an impact on the properties of nanomaterial. For instance, mechanical disintegration of cellulose induces lower mechanical properties of obtained nanocellulose fibers.

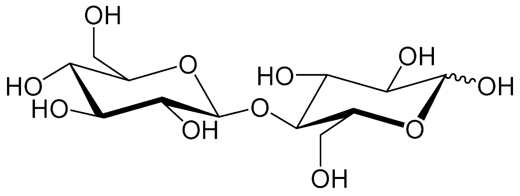


Figure 39: Cellobiose unit, pattern of CNF structure

CNF, and more generally cellulose, are constituted of cellobiose units repetition (Figure 39). The cellulose nanofibrils have generally a length in the range of micrometer and a width of several nanometers [158]. This pattern in CNF structure allows to give good mechanical and barrier properties to materials made with nanocellulose. Indeed, these units agglomerate and, in the overall structure, appear crystalline and amorphous zones. As described in the first section of this report, the proportion of amorphous and crystalline zones has an effect on the mechanical and barrier properties. In particular, oxygen and other compounds transmission through CNF material will occur in amorphous zones, where there is enough space for their path. CNF crystallinity, the proportion of crystalline zones, is around 70% [157]. These fibrils are not rigid and capable of creating a dense compound. Thus, free areas will be reduced, and gas molecules path will be more difficult [159]. This increased density is generated by a well-known phenomenon: hydrogen bonds.

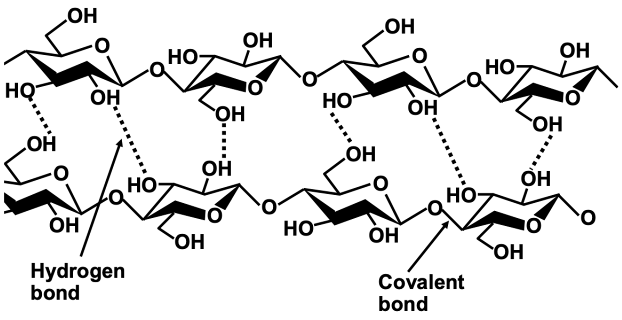
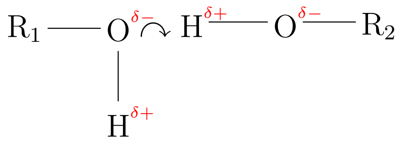


Figure 40: Hydrogen bonds responsible of CNF dense structure [160]

Hydrogen bonds are electrostatic interactions. They occur in between an electronegative atom, in this case the oxygen atom of hydroxyl group, and hydrogen atom, which is partially positively charged. This partial positive charge is due to the oxygen atom, linked to hydrogen atom by covalent bond, which implies an electron movement. The cohesion energy of hydrogen bonds is important and allows to form this dense structure of CNF. These hydrogen bonds and cellobiose units in CNF structure (Figure 40) provide good mechanical and barrier properties, presented in Table 10.



Hydrogen bond mechanism

E (GPa)	UTS (MPa)	Elongation (%)	OTR ($cc.\mu m.(m2.day.kPa)^{-1}$)	MVTR ($g.m^{-2}$)
10	100	5	0.6	234

Table 10: Mechanical and barrier properties of CNF film [159][161]

Concerning oxygen transmission rate value, results are not indicated in barrer unit as in plastic polymers section above. However, PE has an OTR around 100 $cc.\mu m.(m2.day.kPa)^{-1}$ [159]. It shows that gas barrier performance of CNF film is higher than PE film for approximately the same thickness. CNF have also good mechanical properties, interesting for packaging applications. Because of these good properties, CNF alone could be used in packaging sector, but also as coating to improve mechanical and barrier properties of other materials. Because CNF are made with wood, their use as paper coating is coherent in order to increase properties, but also recycling rate of coated papers in packaging sector. Both paper substrate and CNF coating come from wood, which avoids recycling stream contamination. Because CNF coating mainly lies on the top of paper surface, paper packaging mechanical properties cannot be significantly changed. However, it is possible to improve other properties as barrier ones. Indeed, experiments show a 70% reduction of paper OTR thanks to CNF coating [162]. CNF films also provide good transparency. This transparency was measured up to 90%. This low opacity of CNF can be explained by densification of the structure, which provides a low dispersion of light. Transparency is an important property for packaging. For many brand companies, it is important to use clear and transparent packages to allow consumers to see the inside content. However, CNF is not free of defaults. The main one regarding packaging application is vapour resistance. As shown in Table 10, the moisture vapour transmission rate of CNF is higher than plastic alternatives. It is lower than for paper, which has a MVTR around 700 $g.m^{-2}$ as described above, but it is still too high to respect moisture barrier criteria of packaging sector. Nowadays, synthetic polymers used for moisture resistant barrier have a MVTR under 10 $g.m^{-2}$. With a MVTR higher than 200 $g.m^{-2}$, it is difficult for CNF films to compete with the synthetic alternatives. This low moisture resistance is mainly due to what makes the highly dense structure and good properties of CNF highlighted above: hydrogen bonds.

As in hydroxyl groups, water molecules are characterized by polarity in between protons and oxygen atom. Thus, hydrogen bond can be created with hydrogen atoms of other molecules. This is the case with cellobiose units in CNF structure (Figure 41). An oxygen atom of hydroxyl groups will form a hydrogen bond with a proton of

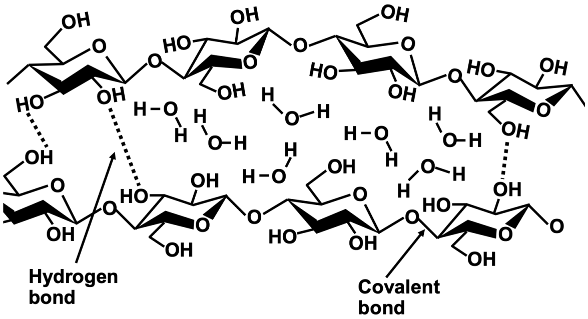


Figure 41: Effect of H_2O molecules on CNF structure organization [160]

water molecules and inversely, reducing cohesion with other cellobiose units [160]. Main problem of this phenomenon is that this moisture weakness has an impact on the other properties of CNF films. Especially, OTR is largely impacted in presence of high humidity content. Because water molecules have an impact on structure cohesion, there is more space created for oxygen transmission. Indeed, starting from 60% in relative humidity, an important increase in OTR with increasing humidity content is observed [163]. The OTR can be multiplied by 20 for high humidity content compared to OTR values for relative humidity under 50%. This default is one of the main default for packaging applications, because it could have an impact on content storage time.

In the aim of reducing the impact of humidity on gas and mechanical properties of CNF films, coating options have been under studies. Several materials were under studies, as coating agent for CNF films, with hydrophobic properties and acceptable biodegradability to keep the sustainable aspect of CNF films. Synthetic plastic coated CNF films could have a lower MVTR, but it will impinge on the recyclability and biodegradability of the final product. By using a binding agent, biodegradable coating based on soy oil increased the moisture resistance of CNF film [164]. Moisture vapour transmission rate knew a 60% reduction with a relative humidity of 50%. This is an important decrease, but not sufficient to reach packaging standards for content protection. CNF film coating has also an impact on transparency of the final product. As explained above, due to low light dispersion of nanocellulose fibrils dense structure, films made with this material have a good transparency. The remaining opacity is created by rough surface, induced by fibers which scatter light. By applying coating on the top surface, single fibrils are shrouded. Thus, CNF film roughness is decreased and transparency improved [165].

5.2.3.2 Modified cellulose

CNF films present interesting mechanical and oxygen barrier properties. Moreover, their good transparency make them good candidate to replace synthetic alternatives currently used by this sector. Furthermore, CNF films dimensions are closer to plastic than paper packaging, and with better properties. The remaining problem with nanocellulose fibrils based materials is its hydrophilic behavior, due to the hydroxyl groups of cellobiose units that constitute the overall structure. This affinity engenders a high MVTR, too high for packaging applications, that can be partly countered through coating agents. Synthetic or biobased, these coating agents have an impact on the recyclability of the product. In the case of synthetic coatings, it could even make it impossible, as for existing plastic coated paper packaging. Hence, it is important to find a way to create a recyclable and efficient coating for CNF film, to make it a serious contender to synthetic plastic film. Recently, modified cellulose coatings were developed. This modification of cellulose provides good moisture resistance and could solve the water vapour resistance problem linked to CNF films. This solution is developed by VTT group in Finland.

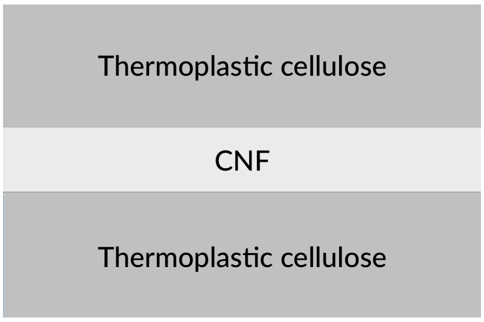


Figure 42: Three layers cellulose material structure developed by VTT group

This technology is based on a multi-layer material, made only with cellulose (Figure 42). CNF constitutes the inner layer, while the two external layers are made with modified cellulose, thermoplastic cellulose. In order to make cellulose become more hydrophobic and resistant to moisture, it is important to modify the hydroxyl groups, that form hydrogen bonds with water molecules. Thermoplastic cellulose was created under this concept: to replace hydroxyl groups by groups with a lower affinity with water. In cellulose structure, each ring owns three hydroxyl groups, which can potentially be substituted (Figure 43).

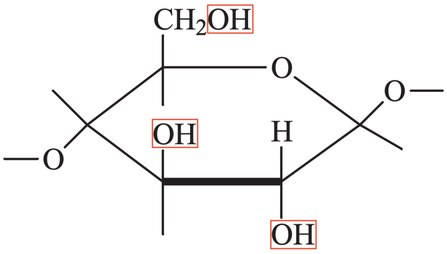


Figure 43: Hydroxyl groups (red) in cellulose structure [166]

The number of hydroxyl groups substituted is called the degree of substitution. In the case of cellulose, its maximum value is then 3, knowing that each ring own three hydroxyl groups. Even if an increase in the substitution degree gives a better plastic behavior to cellulose, and so a lower WVTR, it has to be limited. Hydroxyl groups have a lower steric effect than majority of replacing groups. Steric effect illustrates the fact that each atom, and so molecule, occupies a certain space. When atoms get closer, it implies an energetic cost. This energetic cost is linked to steric effect and the larger the space occupied by an atom, the higher its steric effect. Biodegradation of cellulose involves enzyme attack of the center oxygen atom of cellobiose unit, the oxygen atom bonding the two rings (Figure 39). Enzymes attack of this oxygen atom makes degradation possible, but a higher steric effect could prevent it. Hence, it is important to limit substitution degree to preserve biodegradability of modified cellulose. In order to turn cellulose into thermoplastic cellulose, several parameters can be modified [167]:

- Substitution degree: the higher it is, the closer is cellulose from plastic behavior.
- Length of substituting molecules: by increasing this length, it allows to keep a low degree of substitution, while creating space between cellulose molecules.
- Volume occupied by substituting molecules.

Because biodegradability is an interesting properties that has to be preserved, the second option is consistent. To this end, ester can be used as substituting molecules for hydroxyl groups. To replace hydroxyl by ester groups, acid chloride is introduced. Acid chloride is more reactive than carboxylic acid. In the case of thermoplastic cellulose formation, acid chloride has the following formula: $CH_3(CH_2)_nCOCl$. It leads to the replacement of hydrogen atom in hydroxyl groups by: $-CO-(CH_2)_n-CH_3$. The n parameter, that represents length parameter, can be adjusted to modify the obtained properties of the thermoplastic cellulose [168]. Indeed, it was shown that the higher this parameter, the lower the MVTR of final thermoplastic cellulose film. However, the lower the length of ester groups, the lower the OTR. When ester chain length increases, it creates more space in between cellulose molecules. Hence, it is easier for oxygen to pass through. However, OTR increase stays in the range of CNF. Thus, using long ester chain is better regarding WVTR decrease, while keeping a reasonable impact on OTR. For $n=4$, compared to $n=16$, WVTR is 70% higher. With this substitution of hydroxyl groups, ester thermoplastic cellulose films can achieve a WVTR with around half the value of unmodified CNF films. Some studies found a correlation between n parameter and effect on mechanical properties, but it is still unclear. Mechanical properties of thermoplastic cellulose stay in the range of CNF ones [169].

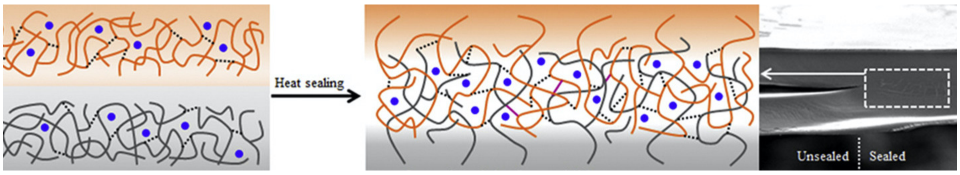


Figure 44: Thermal sealing mechanism [170]

VTT prototype (Figure 42) consists in a three layers material, with two layers of thermoplastic cellulose and one layer of CNF in the middle. CNF layer is used for all the reasons described above, including good mechanical and oxygen barrier properties. As for thermoplastic layers, they provide moisture vapour resistance. In their prototype presentation, VTT announced that thermoplastic cellulose and CNF association pouch could compete with existing materials, including PP and PVC. Their prototype had a WVTR in the range 0.5-10 $g.m^{-2}$. Thermoplastic cellulose layers also provide an interesting property for packaging sector: thermal sealing. As explained in 5.2.1, thermal sealing (Figure 44) is essential for packaging closure and content protection. However, unmodified CNF material cannot achieve thermal sealing. Thermal sealing strength of this multi-layer material was low compared to synthetic alternatives used currently, but it is function of coating thickness. When thickness of thermoplastic cellulose increases, sealing strength increases too. With normalized results, the sealing strength was comparable to classic plastic alternatives, including PE [171].

Thermoplastic cellulose and CNF association acts as a good alternative to current plastics in the field of packaging. Thermoplastic cellulose coating could also be used to coat paper packages, and improve their barrier properties without degrading their recycling rate. But before democratizing the use of this multi-layer cellulose material, it is important to study the different challenges that could exist regarding its production or end-of-use treatment.

5.2.3.3 CNF recycling and challenges

CNF recycling was studied through conventional paper recycling process. This process consists in dipping nanocellulose material into water, before shredding it intensely to separate fibers. The obtained material is then converted into new CNF film. The recovered nanocellulose product had lower mechanical properties and the barrier properties were slightly modified compared to virgin one [172]. However, these properties stay adequate for packaging applications. The reason for this change in CNF properties is due to improper fiber aggregates separation from the initial material, that influences the structure of the recycled one. Moreover, the recycling method did not make use of chemicals. The use of chemical could have improved the properties of the recovered material. Nevertheless, it shows that CNF products are recyclable through classic paper recycling process. Hence, even if the recovered material is not used for CNF production, it could be used for paper production. In the case of an association nanocellulose-synthetic plastic, interesting results were found. The use of CNF as barrier layer in plastic packaging has been studied to reduce plastic production of the packaging sector, avoid multi-material plastics packages and improve properties [173]. Without mentioning the ecological impact, that may be limited, this association had no harmful effects on plastic recycling [174]. Recycled coated plastic had similar properties to recycled virgin one. These results mean that CNF coated plastic packaging made with PE can be recycled in usual PE recycling stream, without impacting the properties of the recycled material.

Nanocellulose materials present good recycling suitability and do not have effect on recycling of other materials when associated. However, despite end-of-use treatment, it is important to study the production cost of such a material. As depicted in 5.2.2, plastic replacing has to be done with sustainable materials in terms of wastes production, to limit littering, but also in terms of production impact. But the use of CNF for packaging sector is only at the beginning of its development. Indeed, it will soon be introduced at industrial scale, thanks to VTT group collaboration with packaging supplier companies. Hence, the large-scale impact of this new product has not been highlighted. Several points for CNF packages production were, or still are, under debate, on ecological but also commercial aspects.

On the ecological aspect, the energy consumption for CNF production is highlighted. As explained above, there are several mechanisms to produce nanocellulose fibrils. The used mechanism depends on the wanted properties and qualities of CNF. It often operates at high pressure, in order to make primary wood cellulose enters homogeneously in the separative device, and requires a high amount of energy. Thus, production of CNF is not energy effective, with energy requirements around 137 $MJ.kg^{-1}$ for median value and up to 234 $MJ.kg^{-1}$ [175]. Comparatively, the energy production for classic synthetic polymer alternatives is lower. In the case of PP and HDPE, the energy requirements are respectively around 86 $MJ.kg^{-1}$ and 74 $MJ.kg^{-1}$ [176].

Material	CNF	PP	HDPE
Energy requirement ($MJ.kg^{-1}$)	43-234	86	74

Table 11: Energy requirements for CNF and polyolefin alternatives [175][176]

Because this high energy requirement is due to high pressure process, in order to get homogeneous cellulose upstream, studies were done to find other ways to achieve it, with a lower energy consumption. At the present time, homogeneization is done with chemicals and proteins first, to reduce process energetic cost. Thanks to this new homogeneization process, the energy requirement for CNF production is noticeably lower. Indeed, it is now in the range 1.8-5.4 $MJ.kg^{-1}$ [175]. This new process consists in using proteins, and more particularly enzymes, which will help to separate cellulose fibers. These fibers form agglomerates which have to be separated to produce nanofibrils [177]. Thus, energy challenge linked to CNF production has been resolved, and protein treatment allows to reduce the production energetic cost below polyolefins one (Table 11).

On commercial aspects, mentioned arguments against CNF use in packaging sector are the maintenance and production cost. Production cost includes expensive compounds used for homogeneization and others. With several processes, CNF production cost was evaluated around 220 $\$.kg^{-1}$ [178]. This price is largely higher than for polyolefins, often under 10 $\$.kg^{-1}$. However, this production cost is depending on a large number of parameters, and it is actually complicate to compare it with polyolefins one. Indeed, CNF is not yet produced at an industrial scale as large as for synthetic alternatives. Though commercial development will take time, nanocellulose packaging could be a good replacing option for current plastics. Nevertheless, it will be complicate to replace all plastic packaging products by CNF ones. Moreover, the ecological impact of this total replacing is not yet known. Hence, it is necessary to improve the existing and main problem of plastic packages end-treatment: recycling.

5.3 Recycling methods

Part 3.5 of this report presented mechanical recycling as the only way to recover materials from plastic wastes. At the present time, it is the only way for plastic materials recycling at industrial scale. Indeed, mechanical recycling is a simple process, low-cost, that provides an environmental advantage. However, this recycling method is not a long-term method in the aim of achieving a closed loop packaging sector. In a first time, mechanical recycling requires sorting of polymers types to avoid stream contamination. Sorting methods being not perfect, it reduces the recycled plastic quality. Then, through this recycling system, plastic wastes can be processed only 6-8 times at most. When this limit is reached, the material becomes useless and is finally burnt or disposed in landfill. Moreover, mechanical recycling plants cannot process 100% of their inputs. For instance, in Veolia plant (Saitama, Japan), among the 21,000 tons of plastic wastes that entered the factory in 2019, only 50% were converted into pellets. The rest was incinerated for energy production or disposed in landfill. The reason for this high percentage of loss is that many plastic packaging wastes are too complicate to sort and recycle through mechanical recycling (cf. 5.1). One way to recover these problematic wastes, and divert them away from incineration or landfill, is advanced recycling. Advanced recycling refers to several methods, more or less developed, but rarely present at industrial scale at the present time. The aim of advanced recycling is to reduce polymer molecules into smaller ones. Thus, it can be called feedstock recycling too. These new molecules can then be used for different applications. Advanced recycling could help to recover rejects and mixed plastic wastes. The different methods will be presented in this part.

5.3.1 Thermochemical recycling

Thermochemical recycling is part of advanced recycling. It allows to transform plastic wastes into raw materials. These raw materials are generally at the bottom of the plastic production process. The two processes presented here are liquefaction and gasification.

5.3.1.1 Liquefaction

Liquefaction, often referred as pyrolysis, is a recycling process whose product is liquid oil. Pyrolysis consists in heating plastic wastes at high temperature, without oxygen, to obtain liquid oil. This oil can then be used to produce new materials in refinery, or burnt for energy production. This system presents similarities with the one described in EfW part (cf. 3.4.2). The difference being that it produces a material and not directly energy. In the case of pyrolysis, plastic wastes, that can be pre-sorted upstream, are sent in a reactor to be processed at high temperatures. Temperatures are mainly in the range 400-600 °C, but depend on the wanted product, waste nature and polymer type [179]. Different types of reactor exist, including fluidized-bed reactor, which principle is similar to fluidized-bed incinerator explained previously. The type of reactor influences the final product. Plastic wastes are not treated in the same way from one reactor type to another. For instance, fixed-bed reactors know mixing problems between wastes and catalyzer, problems that fluidized-bed does not encounter. However, fixed-bed reactors are simpler to compose.

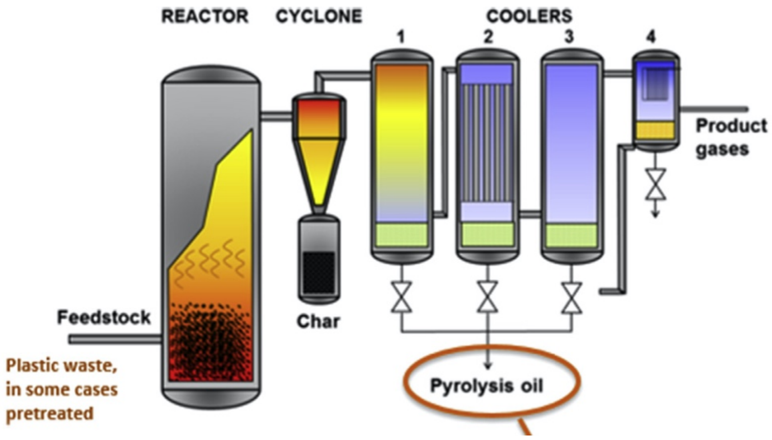


Figure 45: Pyrolysis process [180]

Vapor produced from reactor is then sent in coolers, where it is condensed in liquid oil and separated from remaining gas. Indeed, products of pyrolysis include liquid oil but also gas. Because pyrolysis process is focused on oil production, several parameters have to be controlled, including temperature. In order to keep a high ratio of oil, it is necessary to keep temperature low. However, too low temperatures do not produce qualitative oil. Temperature is an important parameter in pyrolysis and it has to be chosen carefully. The aim of this technology is to be used for mixed plastic wastes recovery. It is currently possible, but the oil yield is lower than with pre-sorted plastic wastes. Indeed, oil yield was observed higher than 90% weight for certain pre-sorted polymers (the remaining 10% being gas and residues). On the opposite, the highest value for mixed plastic wastes was around 50% weight [181]. However, in both cases, pre-sorted and mixed plastic wastes produced equivalent quality oil. Catalysts can also be used to increase oil yield and decrease required temperature. The pyrolysis oil obtained at the end cannot be used directly other than for energy production by combustion [180]. In order to be used by the industrial sector for diverse applications, this oil must be purified. Purification process is done by distillation of the pyrolysis oil. Distillation of pyrolysis oil from mixed plastic wastes at temperatures below 200 °C provides content with properties similar to diesel fuel, except heat value which is lower in the case of distilled pyrolysis oil [182]. Indeed, studies found that pyrolysis oil from plastic wastes could be mixed with diesel in order to power engines. However, percentage of diesel fuel in the mixture has to be increased for long-term application. Moreover, particles emissions, including carbon dioxide and carbon monoxide, are higher than for virgin diesel [183][184].

In the aim of developing the circular economy of plastic, the interest of pyrolysis oil from plastic wastes is not engine power but new plastic production. But this application is currently complicate. As for mechanical recycling, pyrolysis oil quality is suffering from HCl contamination due to PVC degradation. In order to eliminate this compound, temperatures should be increased above 700-800 °C, but then oil yield is reduced. Thus, it is important to realize a pre-sorting, which makes the whole process not economically viable due to high cost of sorting process. At the present time, plastic wastes pyrolysis opponents claim that the whole process is too expensive for too low benefit. According to a study shared by the *Global Alliance for Incinerator Alternatives*, pyrolysis is more polluting than EfW, and only a tiny percentage of pyrolysis oil from plastic wastes is used for new plastics production, the main part being burnt as fuel [185]. Nevertheless, pyrolysis technologies are evolving and could play a bigger role in a near future. BASF company is currently developing packaging products made with pyrolysis oil from plastic wastes. These packaging products could even be used for food applications [186].

5.3.1.2 Gasification

Process for gasification of plastic wastes is similar to liquefaction (pyrolysis) described before. Reactors used for gasification are mainly identical to liquefaction, including fixed and fluidized bed ones. The difference lies in the applied temperature and the reactor environment. As explained above, temperature has an influence on the oil-gas ratio in pyrolysis. In the case of gasification, temperatures are in the range 700-1,300°C [187]. Unlike pyrolysis, the main product of gasification is gas. This gas is extracted from plastic waste by heating in supervised environment. Whereas pyrolysis is realized with absence of oxygen, this is not the case for gasification, with a precise amount of air or steam in the reactor. These environment components are called gasification agents. Gasification agents have a real impact on the final gas composition. The final synthetic gas obtained by this process is called syngas. Syngas has different applications and is composed mainly of methane (CH_4), dinitrogen (N_2), dihydrogen (H_2), carbon dioxide (CO_2) and carbon monoxide (CO). Its properties are mainly influenced by gasification agents and temperature.

To obtain syngas, plastic wastes are heated in presence of gasification agent. The main used agents are steam, air or a mixture of both. However, it is possible to use other gases, including dihydrogen. Because of this agent and temperature, different reactions occur between carbons atoms of feedstock ($C_{(\text{feed})}$) and gasification agent components, allowing feedstock consumption and syngas production. The main reactions are the following ones [188]:

- $C_{(\text{feed})} + CO_2 \leftrightarrow 2\ CO$ (Boudouard reaction)
- $C_{(\text{feed})} + H_2O \leftrightarrow CO + H_2$ (water-gas reaction)
- $C_{(\text{feed})} + 2\ H_2 \leftrightarrow CH_4$ (hydrogasification reaction)
- $CO + H_2O \leftrightarrow CO_2 + H_2$ (water-gas shift reaction)

Used agent influences different properties of the obtained syngas. These properties are mainly syngas heat value, composition in chemical elements and tar presence [189]. Tar is not a wished product of gasification. Indeed, it becomes solid when process is stopped and temperature decreases, obstructing machines. Common removal method consists in using another gas at high temperature after gasification process, to evacuate the created tar. This removal method is time, money and energy consuming. Hence, an efficient method to prevent tar formation is to modify gasification conditions, including temperature and gasification agent [190]. Table 12 summarizes effect of gasification agent choice on syngas composition regarding tar (daf mention indicates that the result is for dry content without ashes), heat value and present elements.

	Heat value ($kJ.kg^{-1}$)	Tar ($g.kg_{daf}^{-1}$)	H_2 (% vol)	N_2 (% vol)
Air	4-8	4-62	11	52
Steam	12-14	61-94	47	0

Table 12: Effect on gasification agent on several syngas properties [189]

These data highlight the impact of the gasification agent on heat value property. Heat value is lower when using air compared to steam. Syngas obtained with steam has a better heat value, but also a more interesting composition for chemical applications. It is free of dinitrogen and has a higher dihydrogen content, which also explains the difference in heat values. Hence, gasification with steam appears to be the best option for both energy production and chemicals production. However, gasification with steam produces a higher content in tar than with air. Tar is problematic for machines in energy production plants, but also for chemicals production through syngas. Moreover, tar production is increased when using plastic wastes compared to biomass as feedstock, which worsen the effect. It also requires more energy for gasification through steam agent. This is due to the necessary energy heat for steam production. For these reasons, syngas produced with steam will be mainly use for chemical production (qualitative) and syngas produced with air for energy production (quantitative). Temperature also has an impact on syngas properties, including heat value and composition, as depicted in Figure 46. Other parameters can influence final syngas, including reactor pressure and mass ratio between feedstock and gasification agent.

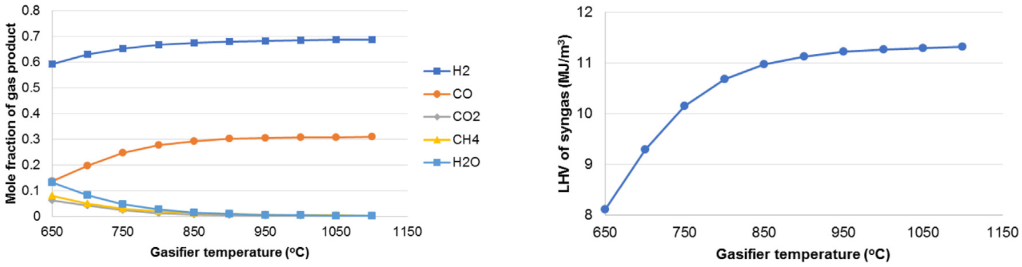


Figure 46: Temperature influence on composition (left) and heat value (right) of syngas obtained from polyethylene wastes and steam [187]

New gasification methods are under development, in order to improve gas quality and yield. One of these methods is plasma gasification. By using a special type of reactor, plasma discharge is used to turn plastic wastes or other compounds into syngas [191]. The advantage of plasma gasification is that it does not produce tar or other gas contaminants due to the high temperatures, which imply intense chopping of feedstock. The obtained syngas has also a better quality, with higher percentage of dihydrogen and carbon monoxide. A recent evolution of plasma gasification makes usage of microwaves. It is mainly the same concept as plasma gasification, but more localized. Instead of using plasma on the whole feedstock, microwaves create localized and small plasma, with brief lifetime, that increases drastically the temperature [192]. Thus, the average temperature of the reactor stays around 1,300°C, while small and local plasma engenders temperature increase that can exceed 4,000°C. One advantage of microwaves gasification is that the intense and efficient treatment of the feedstock allows to process non-conventional feedstock, including hazardous and mixed plastic wastes. Another advantage is the energy economy.

Gasification process is already used as commercial scale, but syngas production from plastic wastes is still rare and under development. Currently, main part of the produced syngas comes from coal or other gases. More than 50% of produced syngas worldwide is used to make industrial fertilisers, but more than 10% are dedicated to methanol production [193]. Methanol is important for plastic production, because it can be used to produce olefins [194], and so help to partly close the plastic loop. As for pyrolysis, gasification process has several drawbacks regarding plastic wastes treatment, that limit its development. The main drawback is shared with pyrolysis: energy consumption. Because gasification requires high temperature in order to convert feedtsock into gas, it consumes a high amount of energy. However, new gasification technologies, including microwaves one, could participate to the energy improvement of this process. Currently, several plants worldwide are processing wastes, including municipal solid wastes, as gasification feedstock. Several plants dedicated for plastic wastes gasification were launched in Japan at the beginning of the 2000's. At the present time, not all are still operational. Closure happened a decade after launch, because of difficulties to manage only plastic wastes as raw materials (transportation cost) and make it economically viable. However, several are still in process in this country, and are part of new recycling development projects.

5.3.1.3 Use of produced char

As depicted above, main products of pyrolysis and gasification are gas and oil, depending on temperature and other parameters. These processes also engender unwanted materials, including tar, that can deteriorate machine systems. Another substance is produced from thermochemical recycling: char. Char is considered as residual matter from pyrolysis and gasification processes. Hence, only limited studies were realized on this product. Char quantity generated depends on temperature, and a temperature increase has for effect to reduce the presence of char while modifying oil/gas ratio [195]. Char can be used for energy production, but this paragraph aims to present another use for this material.

Indeed, Pyrowave company recently patented a process that allows to recover styrene monomers from char. This char is produced from polystyrene wastes, including plastic packaging wastes. The process is a pyrolysis process, prior to oil production, with microwave and catalytic improvement. Temperature is in the range 300-500°C, and microwaves frequency in the range 1,000-2,000 MHz. A compound is added to the feedstock in order to improve its microwave absorption [196]. Thanks to this new pyrolysis process, styrene monomers were produced from char. The weight percentage of styrene monomers in char is up to 90%, depending on the quantity of catalytic compound in char obtained through the pyrolysis process. This technology could also be used for the processing of other types of plastic wastes, including PE and PP.

Both pyrolysis (liquefaction) and gasification could participate, on a large scale, to the elimination of plastic packaging wastes and their reprocessing in valuable materials. These methods are often confined to the treatment of mechanical recycling plastic rejects or contaminated plastic wastes, but supporters insist on the fact that it could be used to process all plastic wastes. Moreover, recent development of these technologies could help to produce high quality material, usable for plastic production. Thus, necessary virgin materials would be reduced and plastic wastes would avoid landfills. They would also engender new material, on the opposite of EfW process. However, current syngas and pyrolysis oil are mainly used as fuel for energy production or other materials than plastics.

5.3.2 Enzymatic monomerization

Pyrolysis and gasification are interesting processes. They allow to obtain, from plastic packaging wastes, materials with quality similar to virgin ones (gas and oil). Nevertheless, these products cannot directly be processed into new plastic packaging. Indeed, they are at the bottom of the production chain, and additional raw materials have to be used. The project of creating monomers from pyrolysis char is promising, but it will stay an annex product of this process, mainly focused on oil and gas production. Hence, with these systems, plastic packaging end-of-use is upgraded, but the concept of closed loop is still distant. In order to close the plastic packaging loop as much as possible, new recycling processes should be created. These processes should draw from polymer formation reaction, and particularly its inverse reaction: monomerization (or depolymerization). These processes should generate monomers as main materials from plastic polymer wastes and operate at lower temperature than pyrolysis or gasification, both energy consuming.

In this context, researches were conducted to find molecules that could participate to the depolymerization process. Several enzymes appeared to have an effect on plastic polymers deterioration, leading to the production of monomers. Enzymes are proteins that act as catalytic compounds in chemical reactions. Each chemical reaction requires a minimum amount of energy to occur. This energy is called the activation energy and its value depends on the reactants and catalysis among others. Enzymes own active sites, where reaction takes place. Through different chemical mechanisms, including reactants orientation, rapprochement and direct participation, enzymes decrease the activation energy of a reaction, thus favoring it. There are different categories of enzymes, depending on the preferred catalyzed reaction. For instance, hydrolases are enzymes that catalyze hydrolysis reactions on their active sites [197]. Hydrolysis reaction was introduced in PLA dedicated part (cf. 3.3.2).

This reaction requires the presence of H_2O molecules to crack other bonds. Certain hydrolases are specialized in the hydrolysis of ester groups, turning these groups into carboxylic acids. These hydrolases specialized in ester groups are called esterases. Inside esterases category, lipases and cutinases can be found. Both were under studies for degradation of PET. From these researches, another category of esterases specialized in PET degradation was highlighted: PETases. At the present time, this enzymatic monomerization is mainly used for PET among synthetic plastic polymers. However, researches are conducted to develop this process for other types of plastic polymers. Indeed, the existence of enzymes realizing polystyrene biodegradation was already highlighted [198].

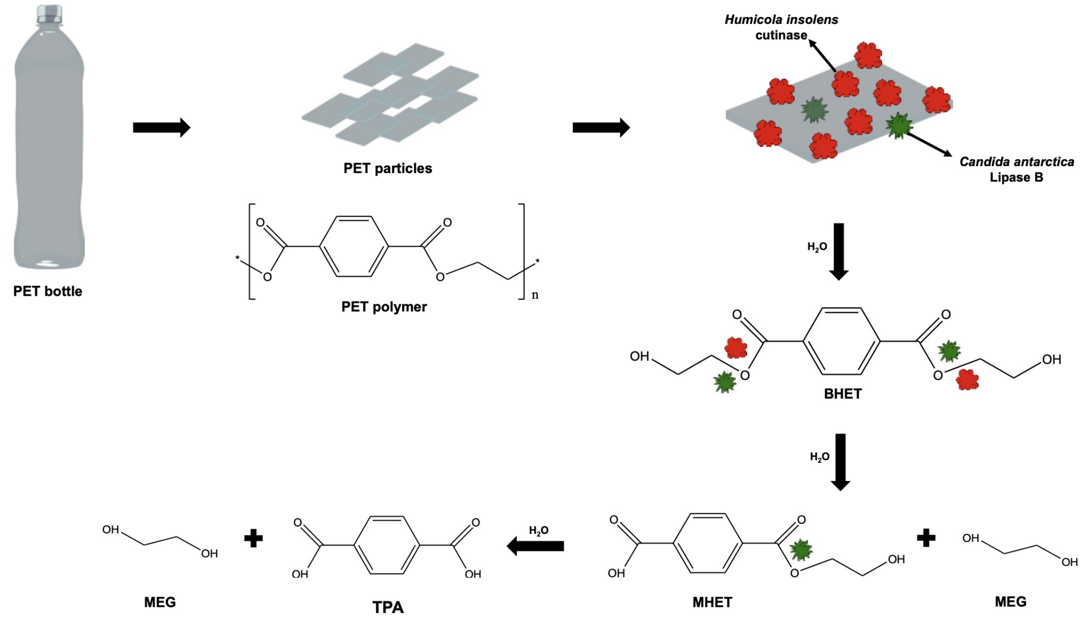
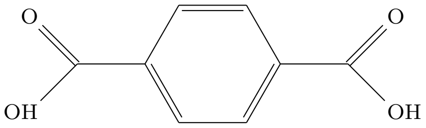


Figure 47: Monomerization mechanism of PET by enzymes [199]

Several studies were conducted in order to find the influence of enzyme type on PET degradation mechanism. In all cases, the final product of this degradation is terephthalic acid, one of the reagents used in dimethyl terephthalate diester formation. As shown in 2.4.1, dimethyl terephthalate is a reagent for PET formation.



Terephthalic acid, sub-reagent of PET

Among esterases, different effects on PET degradation can be observed. As depicted in Figure 47, both cutinase and lipase types degrade PET. But while cutinase easily produces intermediary molecule, lipase realizes directly the entire degradation process and produces a higher amount of terephthalic acid [199]. The differences in degradation path depend on the enzyme type and especially the active sites. These active sites influence the way entering molecule will be affected. Thus, each enzyme type, with proper active sites, catalyzes PET degradation in a different way.

Several other parameters affect the degradation (quality and rate) of synthetic polymers. In the case of PET degradation by enzymes, crystallinity is important. Indeed, enzyme degradation occurs first in amorphous areas (Figure 48), at the expense of crystalline ones [200]. Degradation of crystalline areas is even nearly nonexistent. In these crystalline areas, intense packing of PET chains does not allow enzyme propagation. Moreover, the large proportion of phenyl groups makes difficult PET chains movement. The presence of phenyl groups implies a higher rigidity of the backbone chain. Knowing this, it is understandable that the degradation yield of PET packaging wastes will hardly reach 100%. However, different process conditions can be applied to increase degradation and monomers production yield.

Figure 48: Enzyme degradation occurs preferentially in amorphous zones [200]

At first, PET packaging wastes, including famous PET bottles, have to be shredded in small pieces. PET packaging wastes are turned into microfragments. This shredding allows a higher contact surface in between hydrolases and plastic material [201]. Because enzyme degradation starts from polymer surface, a higher contact surface increases the degradation efficiency. Another parameter improving degradation is temperature. When temperature reaches T_g of PET, polymer chains movement is expanded. Chains movement favors propagation of enzymes in polymer structure and enhances hydrolysis degradation. Thus, it is important to use reaction temperature around T_g, whose value is around 80°C in the case of PET. However, while high temperature, sometimes just below T_g, has for effect to increase chains movement, it also affects enzymes themselves. Indeed, thermal stability of enzymes is limited. When reaching too high temperatures, enzymes lifetime appears to be shorter [202]. Hence, PET degradation is certainly faster, but due to shorter enzyme lifetime, degradation yield is lower. Figure 49 highlights the depolymerization of PET by special cutinase, obtained by plants composting. This cutinase, when used in natural state, allows a 30% depolymerization of PET at 65°C, in three days. After this delay, depolymerization mechanism is stopped. This stop is due to thermal stability of cutinase, because a further addition of this enzyme triggers again the depolymerization mechanism. Nevertheless, researchers succeeded to find the part of cutinase structure responsible for thermal stability. They modified this site in order to increase thermal properties of this cutinase, including melting temperature. Thus, modified cutinase allows a depolymerization yield of 80% at 72°C, in 12 hours (Figure 49). However, thermal stability is still limited and a bigger increase in processing temperature makes depolymerization yield fall below 60%. Quantity of enzyme used can also be increased to reach a higher yield. By tripling the mass of modified cutinase used (at 72°C), depolymerization yield outreaches 90% in the same time, compared to previous 80%.

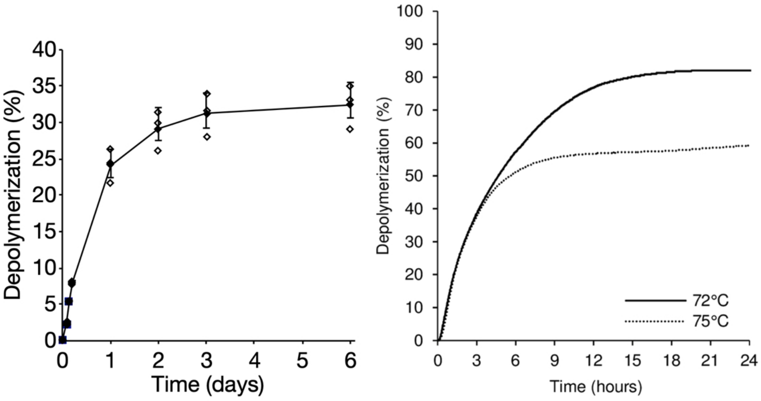


Figure 49: Depolymerization with cutinase at 65°C (left) and modified cutinase with increased thermal stability (right) [202]

Another advantage of this enzymatic approach is that it could solve the problem of many other end-of-use treatments: plastic wastes mixture. In theory, it is possible to combine enzymes, dedicated for different polymers degradation, into an enzyme mixture, and use it to turn mixed plastic wastes into different monomers. This concept of enzymes mixture was already raised for PET degradation [203]. However, this is still theoretical, due in part to the difficulties to find suitable enzymes for polyolefins depolymerization. Enzymatic degradation could also be used to help recycling of laminated packaging films. As introduced above, multi-layer packages are difficult to recycle because of polymers mixture, that could contaminate other recycling streams. Enzymes showed capacity to degrade the internal layer, thus allowing to separate different polymers from laminate packaging [204]. The enzymatic treatment had nearly no impact on the properties or recyclability of external layers. This treatment could then be implanted upstream, in packaging recycling process, to improve multi-layer packaging recyclability.

The process for PET enzymatic monomerization is mainly developed by French company Carbios. Carbios owns the patent for this process and is actually building a plant based on this technology in France. The plant will have an annual processing capacity in the range 50-100 kt. Its objective is to expose the viability of this monomerization process at industrial scale. With this PET enzymatic monomerization, Carbios technology can produce up to 0.2 $kg.kg_{PET}^{-1}$ of terephtalic acid in several hours, in addition of ethylene glycol [205]. Moreover, quality of the obtained product is similar to virgin one.

6 Conclusion

Recently, worldwide plastic consumption is expanding. This trend should continue, until reaching dramatic level in several decades if no action is taken. Without talking about population increase, the reasons behind this large plastic use are numerous, but one particularly stands out: plastic is efficient. Plastic is not only cost efficient, with a low production price, but also efficient because of its properties. There is a large diversity among the types of polymers used for plastic production. This diversity translates into properties diversity. Thus, it is possible to obtain polymers with high gas barrier (PET), low density while protecting content (EPS) or even polymer types that can be modified through additives for a further diversity (PVC). This wide panel of properties makes this material the best choice for packaging applications. Moreover, plastics are mainly durable, which allows their use in many environments and a long content protection, in case of plastic packaging. This last property is certainly its main drawback: plastic is hardly eliminated. When leaking in the environment, it takes hundreds of years to degrade, due to a poor, next to nil, biodegradation. Among the plastic wastes the more likely to leak in the environment, including oceans, plastic packaging wastes are in first position. Indeed, their small size and ephemeral nature make them difficult to manage, and they easily escape the wastes management system. In order to avoid this leakage, and reduce the impact of their increasing production on resources and environment, different end-of-use methods are in place. Although a large amount of these packaging wastes are still sent to landfills, several methods were created over the last decades to produce usable materials from them. Main methods include production of energy, through Energy-from-Waste plants (thermal recovery), and downgraded plastic materials, through mechanical recycling. However, both of these methods face limitations. Above all, hazardous emissions and limited impact are spotlighted. Furthermore, large diversity in plastic packaging compositions creates difficulties for the treatment of these wastes. Even with sophisticated sorting methods, including NIR and optical ones, streams contamination is an inevitable consequence. Hence, it is difficult to obtain recycled plastic materials suitable for packaging use. Mechanical recycling is a low-cost method, that allows to recover part of the plastic wastes into new products. Moreover, between 25 and 50% of the plastic wastes that transit through mechanical recycling plants cannot be processed, because of contamination or technical impossibilities. On the other hand, EfW plants are criticized for ducking plastic wastes problem, and their emissions are under debate. Nevertheless, it is an efficient way to process and eliminate mixed plastic wastes or contaminated plastic packaging and avoid landfilling. Moreover, plastic wastes appear to be interesting feedstock for energy production, with a heat value around 35 $MJ.kg^{-1}$ in average (compared to coal, around 25 $MJ.kg^{-1}$). In order to solve the plastic packaging problems, actions need to be taken. These actions have to be taken by different entities, from companies that create and sell these packages, to governments that can use their authority to limit or prevent challenging packaging. Innovations, in the fields of materials or recycling technologies, could also participate to this sector improvement in a near future. CNF-based packages from VTT or enzymatic recycling seem promising, but still at the beginning of their implantation. Whereas less recent plastic-coated paper and monomaterial plastic packages, and pyrolysis (oil and gas) are already in place. All these innovations could reduce plastic wastes pollution, while reducing required resources through recycled materials. However, to become successful, it should be accompanied by behaviour changes, in order to reduce the amount of packaging used by consumers.

7 Discussion and recommendations

The aim of this section is to share thoughts and interrogations, that did not worth to be included above. Several aspects will be discussed, including packaging reuse and plastic replacement. Recommendations will also be included. These recommendations concern different entities linked to packaging sector. They were created and proposed by the author of this paper only.

Regarding plastic replacement, it is important to evaluate the ecological impacts of alternatives before democratizing their use. Plastic material has numerous disadvantages, but its impact on the environment is well-known. Thanks to studies from organisms, associations or scientists, there is nearly no unknown impact, whether on marine litter, resources consumption or emissions. This is not the case with alternatives. Glass, paper or metals are certainly more sustainable in the collective imaginary, but it is good to remind that they also have an ecological cost. Glass requires a high amount of energy, because of high processing temperatures, and can break during transport. Moreover, for a same package, the amount of material is noticeably superior for glass compared to other materials, creating more transport emissions and wastes at the end. Paper also requires a higher amount of material, but it is closer to plastic packaging. However, paper barrier and mechanical properties are insufficient, and the existing paper packages for plastic replacement have to be coated with plastic. The proportion of plastic is low, but it makes packaging recycling impossible in paper stream. In addition, even by using well-managed forests, replacing all the current SU plastic packages could engender a large deforestation. The amount of water used for paper production is also high compared to plastic. Above all, glass, paper or metal packaging wastes are also concerned by contamination, reducing recycling options. A life cycle analysis has compared ecological impact of different materials for packaging application. Results showed that plastic has the lower impact in many categories, including energy and water consumption, wastes production and global warming [206]. As it was already explained in the above content, life cycle analysis is an efficient tool, but results do not have to be taken as absolute truth. Indeed, it mainly depends on databases used and parameters taken into account. The aim of this paragraph is not to promote plastic packaging, but raise awareness about sustainable alternatives sold by companies.

This introduces the second point of this section: companies responsibilities. With the collective consciousness awakening about plastics, thanks to NPO and documentaries that highlighted the ecological impact of this material, many consumers started to ask for alternative materials. Thus, many brands decided to change their packages from plastic to other materials. However, this change was often done without deep studies on its ecological consequences. By offering alternatives to consumers because it is actually a better selling argument than plastic, companies perform greenwashing. Greenwashing is definitely not a good supporter of packaging sector sustainable development. As a chain official admitted for Green Alliance: "We are aware that [by switching from plastic to other materials] we may, in some cases, be increasing our carbon footprint." [207]. Because companies place packaging on the market, packaging that will inevitably engender waste, they have to study upstream the environmental impact of their products. Thus, companies should develop sustainable materials in packaging, including recycled content, mono-material plastics or innovative materials when relevant. Moreover, concessions have to be made about packaging design to improve recycling. The best way to do that is to follow recycling guidelines created by organisms, including Recoup and Citeo.

Companies should also track their packages, from the creation to the end-of-use. Deposit and refill are good options for this purpose. Terracycle is a company whose system is now implanted in several countries. Its mission is to collect wastes that encounter recycling difficulties. By creating a partnership with Terracycle, companies can improve their packaging collection and create a dedicated recycling path for difficult products. For instance, Lipton collaborates with Terracycle in order to collect and recycle separately its tea capsules. Collection can be done thanks to deposit points or privately, by sending them directly to Terracycle [208]. By creating different collection paths, dedicated to precise packages, recycling is facilitate and contamination reduced.

Collection improvement will be possible thanks to companies involvement, but it will also implicate consumers. Indeed, companies can implant numerous partnerships and give precise indications on their packages, but disposal in the good bin, in the good way, depends at the end on consumers. Even little gestures can participate to end-of-use treatment improvement. According to Ariake Kogyo plant manager, two gestures are particularly important: packaging rinse and sorting rules respect. The first gesture allows to eliminate part of the packaging contamination, responsible for low recycling rate of these plastic wastes. The second gesture regroups different actions, including packaging parts separation and placement in the correct bin. When doubting about the correct bin for a package, it is better to place it in the garbage bin than in the wrong recycling bin, where it could contaminate the whole recycling stream. For instance, contaminated plastic films are rarely recycled, due to dimensions and contamination. Thus, by placing them into the recycling bin, stream contamination of other streams can occur. But the best action regarding packaging, that can be done by consumers, is simply to reduce the amount of packaging used. The best package for the environment is the package that is not produced. This point also concerns companies, that need to reduce the amount of material used for their packages. However, when loose shopping with bulk products is available, consumers should favour this option. In order to transport and store these products, reuse of old packages is a good option, including thin plastic bags and jam glass jar. Finally, consumers should inform their entourage of the actual concerns about packaging, and develop personal interest for this ecological topic, by reading specialized papers as Recycling Today or Packaging News. Nevertheless, it is relevant to notice that consumers are more and more aware of packaging impact. According to surveys, around three out of four consumers would be ready to pay more for more sustainable packaging (15,000 interviewees, Netherlands) [209]. In the same time, 60% would be ready to break their habits and buy products of a brand that uses less packaging than another (2,000 interviewees, UK) [210]. However, only 83% admitted to read and take notes of the sorting instructions on the package (4,000 interviewees, Europe) [211]. This percentage should appear as high, but it means that potentially 17% of European people do not sort or badly, which could have bad consequences on the recycling process. Consumers must become aware of the sorting importance.

But consumers awakening is also based on governments involvement. An important part belongs to consumers' implication, but a general and efficient regulation has to be implanted by governments. Recently, countries started to ban SU plastic packaging, especially SU plastic bags. More should follow, including cutlery, cups and straws, as it was already done in France (cf. 4.4.4). However, no exceptions should be done for bio-based or biodegradable plastics [212]. Indeed, they do not have a proper recycling stream and only degrade in special conditions, not in nature. In order to develop the recycling market, governments should establish rewards for companies using recycled content. In a stricter way, they should establish taxes on products with packaging made of virgin material. This point does not concern only plastic, but also paper and other alternatives. Recycled content prices are still high, higher than virgin materials in the case of plastic due to oil barrel price drop.
Governments should also develop sorting guidelines for population and improve collection system. In UK, each year, paper recycling stream is highly contaminated by plastic packages [213]. This is due to mix collection system for recyclable wastes. Hence, plastic and paper packages end in the same bin, and one can leak in the recycling stream of the other. In the present time, sorting methods are efficient, but still have limitations. Thus, it is important to separate collection methods, as it is the case in Japan (Figure 13). But this separation system requires a higher involvement of the population to be relevant (cf. 3.1.1). However, wastes sorting is still considered as a civic act. In order to make this separated collection system a success, sorting must become an obligation.

In this paper, several materials, recycling methods and innovations were presented. However, it is important to live and act with the current infrastructures. CNF packages and enzymatic recycling could be important and democratised in the future, but they are still in development. In the present time, it is neither possible to rely on them, nor wait for them. Indeed, plastic packages leak in the environment every day, or end up in landfills, and immediate and efficient methods are urgently required. Knowing this, the best option is maybe to use material recycling as much as possible, and use thermal recovery for the remaining plastic packaging wastes that cannot be recycled. This method is applied in Switzerland, country with a landfill ban for burnable wastes. Obviously, EfW is not a sustainable and long-term solution, and it should not replace or delay other recycling methods development, but combined with current material recycling methods, it could help to tackle the actual plastic packaging problem.

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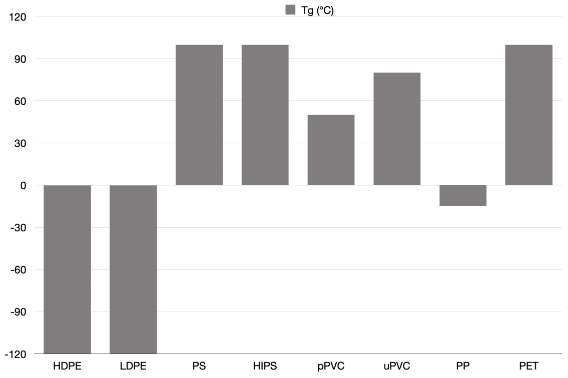
8 Annex



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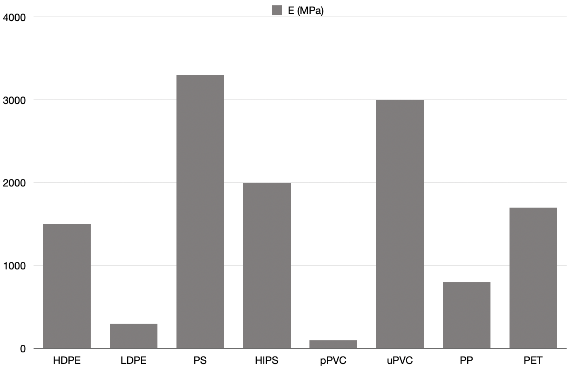

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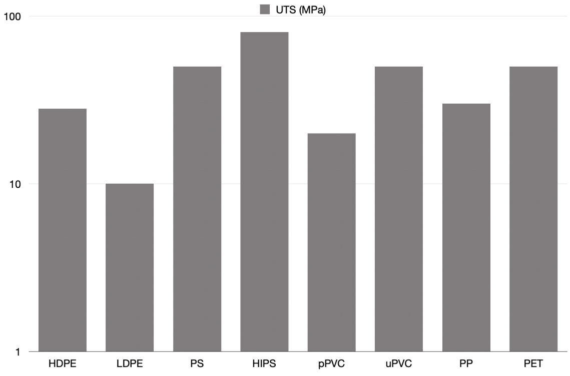

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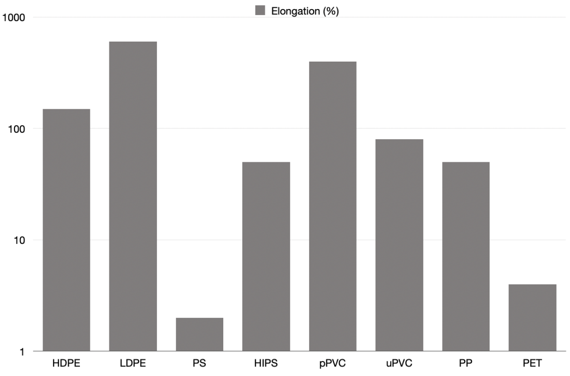

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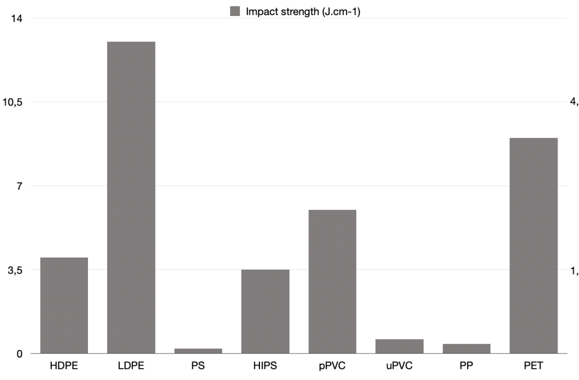

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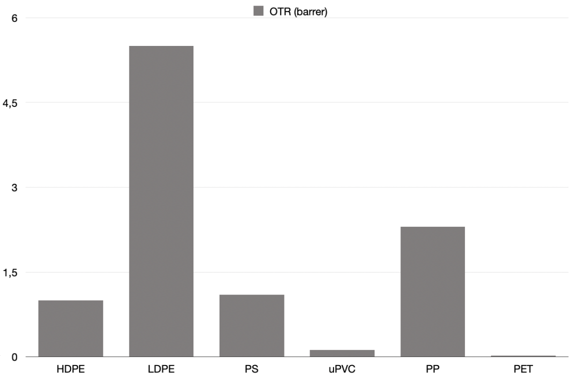
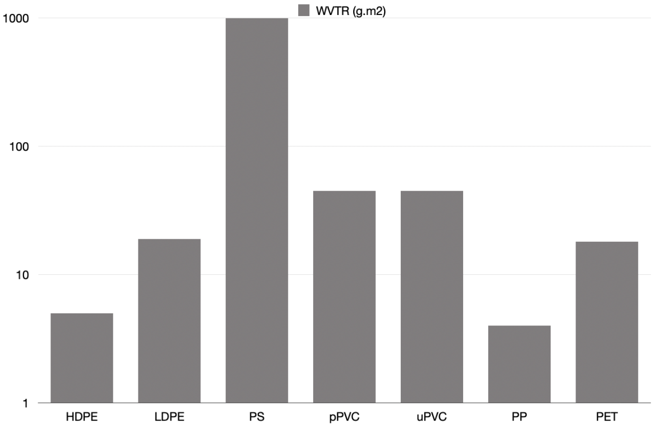


OTR comparison



WVTR comparison