AN INTRODUCTORY REVIEW

MECHANICAL RECYCLING OF POLYMERS FOR DUMMIES

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Authors' introduction

It's a bit unusual perhaps, to name this introductory review 'Mechanical Recycling *for Dummies*'. The text provides an accessible introduction to the technical aspects of mechanical recycling; it is aimed at (polymer) engineering students, needing a general introduction. Or at polymer scientists, wanting to explore the world of recycling and likewise, at recyclers wanting an insight into why polymers are so damn complex to recycle. None of these target groups are dummies. But all of them are out of their comfort zone with at least one section of this review. Recognize yourself in this profile? Then go ahead, read. Recycling (mechanical or otherwise) of polymers is a complex matter, which really does require a multidisciplinary understanding to do well. We have chosen to go broad rather than deep in terms of content. We have selected to just share instead of publishing in a journal, so that anyone can access the text. If you would like to this use educationally, share it within your company or network, feel free. But please, do remember to make the proper references to us as authors. Proper courtesy will keep us encouraged to provide you with more of these open papers.

Abstract

This article will give you a comprehensive overview of the recycling of polymers with a strong focus on mechanical recycling. Starting with an overview of basic waste management hierarchy, the manuscript continues with the principles and scientific challenges associated with mechanical recycling. Furthermore, different industrial technologies are highlighted focusing on post-consumer polyester (PET) bottle-to-bottle recycling, solid plastic waste (SPW) from post-consumer packaging waste and SPW from waste from electrical and electronic (WEEE) equipment. In addition, various additives commonly found in or added to recycled polymers are discussed.

Keywords

Recycling; Mechanical Recycling; Polymers; Solid Plastic Waste

1   INTRODUCTION

Plastics combine some inherent qualities like low density, durability, low cost, good processing capabilities and corrosion resistance making them usable in a wide application range [1], [2]. The production of plastics has risen from 1.5 million tonnes in 1950 to a staggering 348 million tonnes in the year 2017 (64.4 MT for Europe), leading to a total world production of 9 billion tonnes of plastics in the last 65 years. After China, the biggest annual producers of plastics (29.4% of total production of thermoplastics and polyurethanes), are Europe and NAFTA, both good for respectively 18.5 and 17.7% of the global amount in 2017 [3]. The plastic challenges associated with these number involve mainly climate change measured in the carbon footprint and the

plastic pollution causing enormous plastic leakage into the oceans [4].

Plastics possess a large potential to improve their circularity because of low recycling rates, therefore CO_2 gains could be substantial. Mechanical recycling i.e. cleaning, re-melting and upgrading of plastic waste produces less than 20% of the CO_2 emissions associated with making new plastics [5]. In the European Union, the potential for recycling plastic waste remains largely unexploited . Reuse and recycling of end-of-life plastics remains very low, particularly in comparison with other materials such as paper, glass or metals [4].

This article gives a comprehensive overview of the mechanical recycling of polymers. It commences with an overview of the hierarchy within waste management, followed by the principles and scientific challenges associated with mechanical recycling. Furthermore, different industrial mechanical recycling technologies are highlighted focusing on post-consumer poly(ethylene terephthalate) (PET) bottle-to-bottle recycling, recycling of solid plastic waste (SPW) from post-consumer packaging waste and recycling of SPW from waste from electrical and electronic equipment (WEEE). In addition, various additives commonly found in or added to recycled polymers are discussed.

2   SOLID PLASTIC WASTE (SPW)

2.1   Origins of plastic waste

The overall plastics market is dominated by the 'plastic big 5', namely polypropylene (PP), polyethylene (PE), polyvinyl chloride (PVC), polystyrene (PS) and PET, who together make up around 70% of all manufactured plastics worldwide. In Europe, the dominating markets for these plastics are packaging (40%), building and construction (20%), automotive (10%), electronic and electronical (6%) and agriculture (3%). All other markets combined (consumer and household goods, furniture, sport, health and safety) make up the remaining large of 20% [3]. An overview of these markets and their typically used polymers is shown in Figure 1.

Since competition for natural resources increases globally, markets such as plastics production are becoming more vulnerable to access and prices [6].

The European economy is currently experiencing a strong drive towards a focus on sustainability, both in production and consumption of products. European countries have evolved over

the last two decades from a focus on disposal methods to a greater focus on prevention and recycling. Recycling is considered a major waste reduction strategy, as part of the 4Rs strategy: reduce, reuse, recycle (materials), recover (energy) [7]. Recent pivotal documents include the European Commission's Circular Economy Package [8] and the New Plastics Economy report by the Ellen Mac Arthur Foundation [9].

Most of the plastic waste is generated by households and (food) packaging waste in turns dominates this waste stream. In 2010, the average European generated 513 kg municipal solid waste (MSW). Generation of municipal waste per capita has declined slightly from 2004 to 2012, but it is clearly better managed now than ten years ago. Countries that have developed efficient municipal-waste management systems generally perform better in overall waste management. The number of countries recycling and composting more than 30% of municipal waste increased from 11 to 17 out of 35, and those landfilling more than 75% of their municipal waste declined from 11 to 8 [10].

Recycling rates are steadily increasing. In 2016, 27.1 million tonnes of post-consumer plastics waste ended up in the official waste streams. 72.7% was recovered through recycling and energy recovery processes while 27.3% still went to landfill. This trend of increasing recycling and recovery rates and decreasing landfill is shown in Figure 2 [3].

There is a strong legislative drive to continuously increase recycling rates for plastics, especially given the ambitious targets set in the recent Circular Economy Package [9] and its subsequent action plans, which foresee a common EU recycling target of 65% of all packaging waste by 2025 (and 75% by 2030), including a 55% recycling target specifically for plastics packaging.
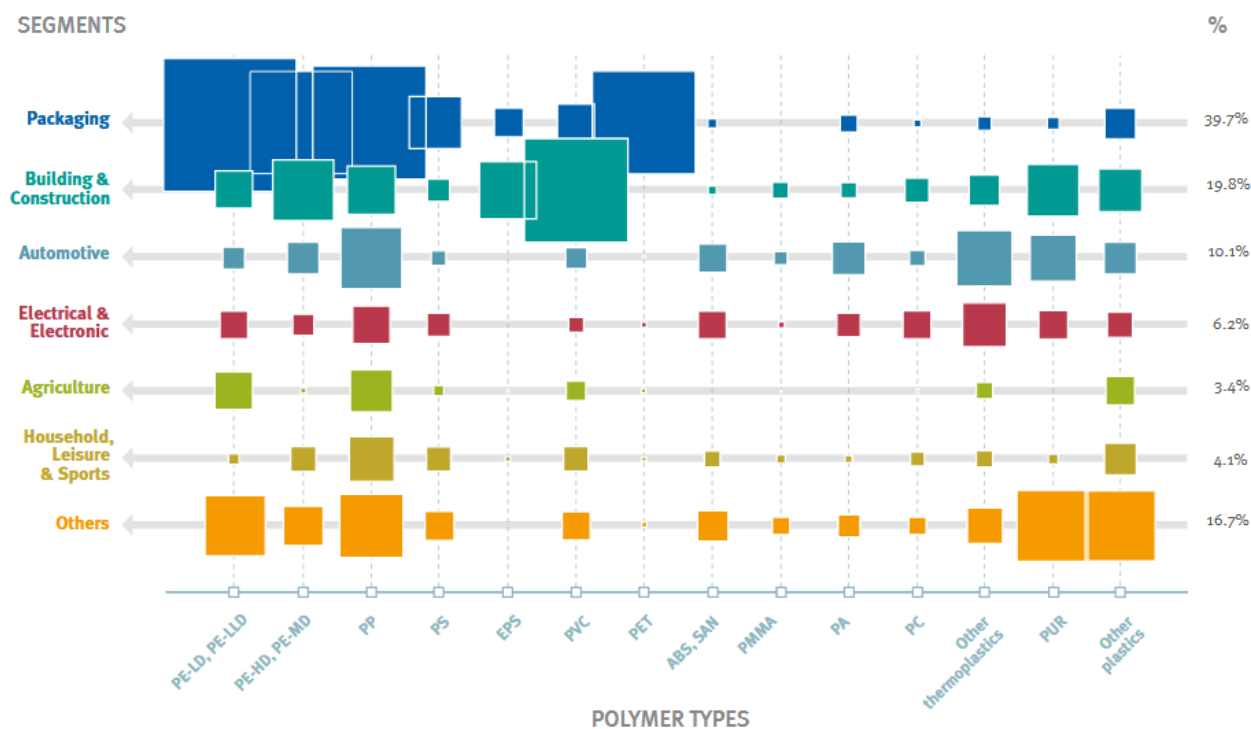
SEGMENTS

Packaging — 39.7%
Building & Construction — 19.8%
Automotive — 10.1%
Electrical & Electronic — 6.2%
Agriculture — 3.4%
Household, Leisure & Sports — 4.1%
Others — 16.7%

POLYMER TYPES

PE-LD, PE-LLD | PE-HD, PE-MD | PP | PS | EPS | PVC | PET | ABS, SAN | PMMA | PA | PC | Other thermoplastics | PUR | Other plastics

FIGURE 1. Plastics demand by polymer and market segment (2017) [3]

2006-2016 evolution of plastic waste treatment (EU28+NO/CH)

Recycling +79%
Energy recovery +61%
Landfill -43%

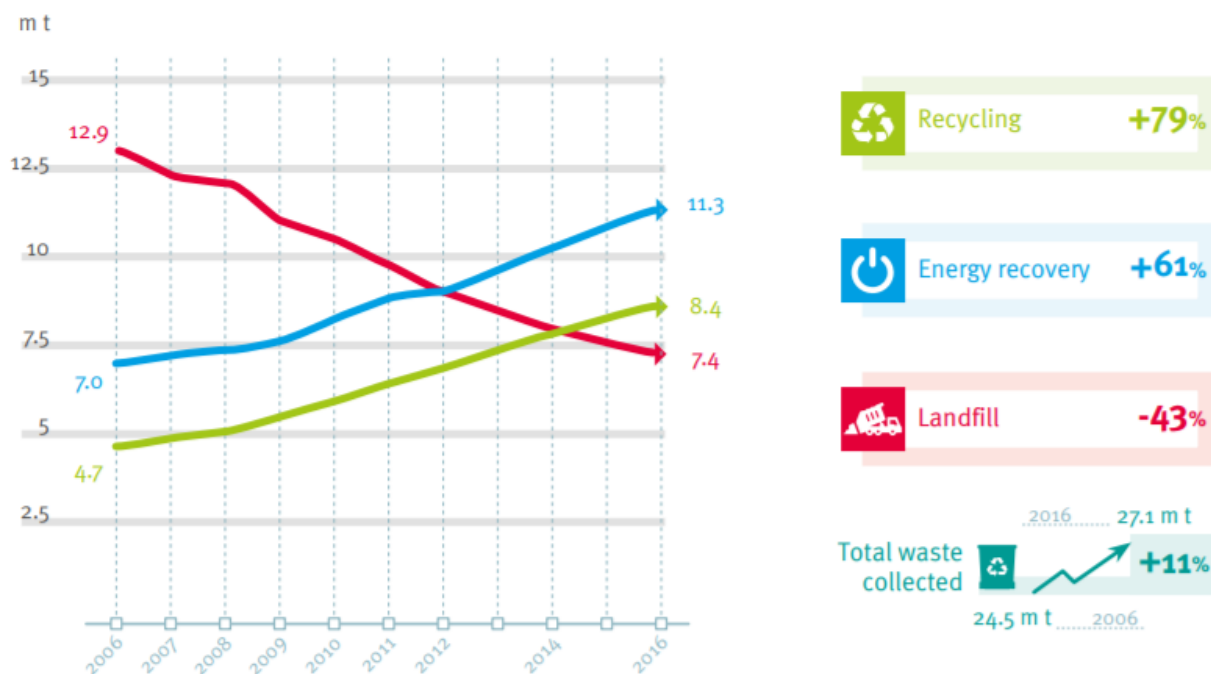Total waste collected: 2016 27.1 m t +11%, 24.5 m t 2006

FIGURE 2. 2006-2016 waste treatment evolution in Europe; green: recycling; blue: energy recovery; red: landfilling [3]

2.2   Waste Management Hierarchy

Like other waste streams, plastic waste has to be coped with in the most sustainable way. The waste management hierarchy in Figure 3 presents a priority list of practices to process waste streams. The list prioritizes the different waste handling methods from least environmentally friendly to most sustainable [11]. Prevention can be found in the uppermost position of this waste management hierarchy. 'Prevention is better than cure' is the primordial objective in waste management, because no collection and processing of the waste materials are necessary. The second most desirable option is re-use, in which the waste materials or products do not undergo structural changes, and are used again for the original purpose.

However, it is inevitable that extensive amounts of plastic waste will still come into existence. Therefore, the focus has to be in the recycling of these waste streams with a view to closing loops and grow further into a 'circular economy' way of thinking. The recycling pathway, either mechanical or chemical, leads to secondary raw materials which can be used for their original or other purposes. When the previous methods cannot be applied, energy recuperation through incineration is the preferred option. Landfilling is the last resort and has to be avoided at all times.
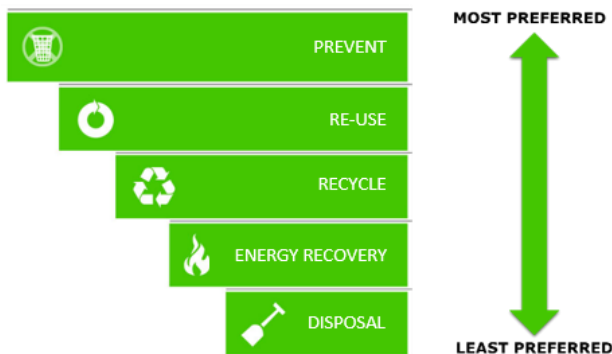

FIGURE 3. Adapted figure from [12]: The Waste Management Hierarchy

2.2.1   Prevention

Prevention is defined [13] as the measures taken before a substance, material or product has become waste, that reduce either (i) the total quantitative amount of waste generate, (ii) the negative impacts of the generated waste on the environment and human health or (iii) the content of harmful substances in materials and products.

Different measures can be undertaken to prevent new waste. Extension of the lifetime of a product can be seen as prevention effort. Through material quality, better and/or modular product design in which spare parts are foreseen, product lifespan can be prolonged. Better product design, involving a reduction (e.g. thinner product or optimised volume/weight ratio) in material use, also reduces the total plastic waste generated. Furthermore, the substitution of plastics by other materials (paper, wood, glass, metal, etc.) can lead to a smaller amount of plastic waste, however, life-cycle assessment studies should be performed to be able to choose the most sustainable material. Finally, the design for recyclability principle will contribute to the prevention of plastic waste materials. Besides lowering the amount of plastic used, the focus also has to be put on reducing hazardous compounds in the plastic production [14].

In order to develop a product with the lowest possible impact on the environment, all stages in the product's life cycle have to be considered. An optimal eco-design of plastic products will have a positive influence on its recyclability (at end-of-life) and the degree to which they can incorporate recycled materials (at start-of-life) [15]. The European Union strongly focuses on growing towards a Circular Economy and sets out directives for its countries. The Eco-design Directive (2009/125/EC) strongly advices the Design for Recycling principle, in which products are being developed regarding a straightforward recycling. Efficient material use and easy dismantling of the diverse materials are the key measures of this principle. However, in this approach, the Design for Recycling strategy only concentrates on the product's end-of-life. Hence, it is also essential to aim attention at the design at the product's start-of-life. Therefore, Design from Recycling [16], [17] is a complementary counterpart of the Design for Recycling principle. Recycled polymer material are matched with potential products based on their strengths and weaknesses.

2.2.2   Re-use

Re-use involves products which have been designed and produced to fulfil a minimal amount of rotations over their life span. Mostly these products are selectively collected, reconditioned and reprocessed for the same purpose. The best known example is the multi-trip plastic crates and pallets. Other examples include refillable drink bottles and reusable plastic containers. Products

sold through second-hand stores (clothing, toys, electronics, etc.) are categorized under re-use.

In order to let prevention and re-use succeed, the awareness of the consumer related to the exhaustibility of earth's resources, has to be (further) elevated [15]. For example, consumers have to recognize that the consummation of single-use grocery bags is unacceptable.

2.2.3 Recycling

a. Mechanical recycling

Mechanical recycling involves only mechanical processes (grinding, washing, separating, drying, re-granulating and compounding) [15]. The obtained recyclates can replace virgin plastics in the fabrication of new plastic products. Common processing techniques after re-melting are injection moulding, extrusion, rotational moulding and heat pressing [18] [19].

This technique is only applicable on thermoplastic materials, as thermosets will not re-melt. Examples of mechanical recycling of post-consumer plastics waste:

➤ Collection and grinding of sorted, clean PP crates and blending of the regrind with virgin polymer to mould new crates;

➤ Collection of low density polyethylene (LDPE) films used in agriculture and industrial packaging, pre-washing, grinding, washing, separating, drying and melt-filtration/re-granulation and processing into refuse bags;

➤ Collection and sorting of PET bottles used for drinks packaging, grinding, washing, separating, drying and processing into polyester fibres, sheets or containers[6].

As mechanical recycling is the most ubiquitous in today's industry, this will form the focus of the remainder of this review.

b. Chemical recycling

Also called thermochemical or feedstock recycling, chemical recycling involves mechanisms in which the collected plastic waste is chemically degraded into its monomers or other basic chemicals. The processes involved are amongst others hydrolysis, pyrolysis, hydrocracking and gasification [15] [19] [22]. The output may be reused for polymerisation into new plastics for the production of other chemicals or as an alternative fuel [23].

Although various techniques have been successfully established in the promising field of feedstock recycling, the industry suffers from high investment levels, high energy consumption and high input levels, making only very large plants economically viable at the current time [24].

c. Energy recovery

Incineration of polymers releases the caloric value of the polymers and is also considered as a recycling process. It is a common and highly practiced method for waste reduction and energy recovery [19], often used upon highly contaminated or complex polymers waste streams, such as medical waste and hazardous-goods packaging. While energy recovery unavoidably terminates the lifecycle of a polymer product, it remains preferable to landfill.

3 MECHANICAL RECYCLING OF POLYMERS

3.1 Introduction

Plastic waste can be recycled in a different ways depending on types of polymers, product and packaging design, if the products consist of the single polymer or mixed polymers [7]. Mechanical recycling is one of the most common methods for recycling of thermoplastic polymers such as PP, PE and PET [15, 25]. This process implies collection, sorting, washing and grinding of the material [15], [20]. A schematic overview is presented in Figure 4.

Collection and sorting are discussed separately below. Further, washing of products is a mandatory step for removal of food residues, pulp fibres or adhesives. There are various techniques to remove residues, e.g. via wet by water or dry cleaning of the surfaces through friction without using water [7]. Afterwards, the size reduction from products to flakes via grinding is the last step in mechanical recycling. The compounding and pelletizing can be the optional reprocessing of the flakes into granulate due to easier work for converters [15].
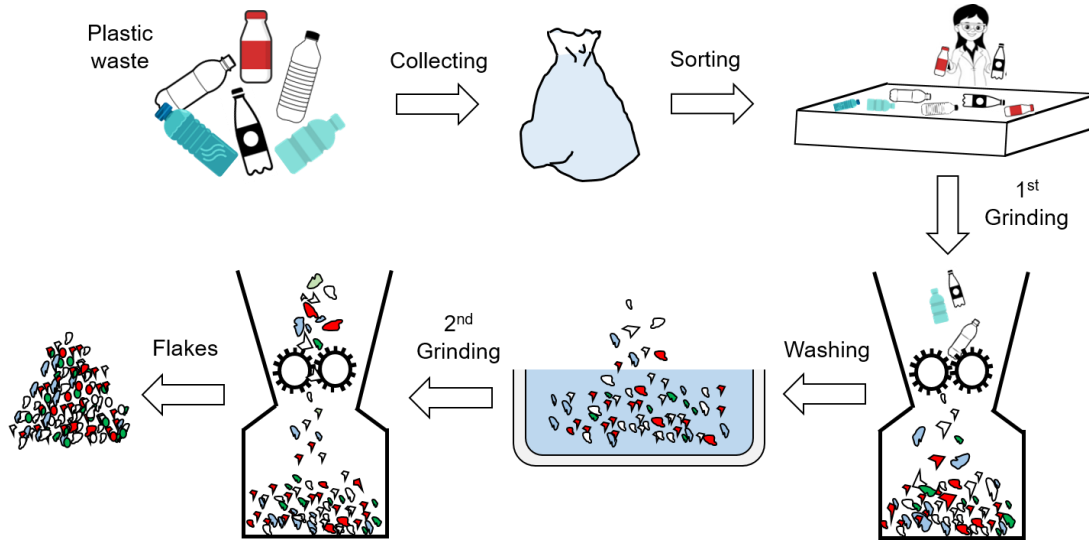
FIGURE 4. Scheme of the basic principal steps in a mechanical recycling process

3.2   Collection

As the first step in the value chain of mechanical recycling, collection systems play a crucial role in the conversion of waste raw materials into new plastic products. Collection schemes should be optimised, on the grounds that they determine the composition of the waste streams and accordingly the downstream procedures, like the pre-treatment, separation and recovery operations [21]. These collection operations should be cost-effective and, approved and supported by waste owners in order to counteract landfilling. There are four main collection methods for plastic packaging waste: kerbside, drop-off, buy-back and deposit/refund programs [22]:

➤ Kerbside collection is the most widely accessible collection method. For civilians, it is the most convenient procedure to participate in, what results in high recovery rates. Residents are requested to separate potential valuable recyclables (plastic, paper and cardboard, metals) from their household waste, commingled or not, into special receptacles or bags.

➤ In drop-off recycling, different containers for designated materials to be recycled, are placed at central community places. Such collection programs often suffer from low or unpredictable throughput.

➤ Buy-back centres, mostly run by private companies, purchase recyclables from consumers. These centres impose specifications to the recyclable waste materials, which makes the contamination level low.

➤ Deposit/refund programs imply the refunding of a deposit when plastic containers are returned to the appropriate redemption centre, or to the original seller.

Besides the municipal plastic waste fraction, proper collection schemes must be supported/designed for the acquiring of valuable raw materials in electrical and electronic appliances, end-of-life vehicles and plastic agricultural films. Furthermore, it will be important to harmonize the different existing collection systems. In addition to the plastic waste collection for individuals, organized sorting and collection of plastic waste from companies is extremely important. Good waste management of plastic industrial waste greatly improves the uptake rates of potentially reusable and recyclable materials. Often companies are rewarded with incentives.

3.3   Sorting

The incoming waste streams consists of mixed plastics of unknown composition and is likely contaminated by organic fractions (such as food residues) and non-plastic inorganic fractions (metals, wood, paper, …) [23]. Cleaning, regrinding and sorting of the waste stream will be necessities in the light of qualitative products through mechanical recycling. An optimal sorting plant knows four different stages to separate the incoming plastic waste streams [24]. First of all, the non-plastic contaminations (metals, wood, paper, etc.) must be removed. The plastic fraction has to be separated in rigid and non-rigid (e.g. foils) components. In order to obtain a good recycled product, the plastic waste materials preferably have to be divided into

coloured and non-coloured (transparent) fractions. At last, the different plastic types have to be sorted out.

In the first place, metals have to be removed from the waste stream, as these can damage the recycling plant's machinery. Ferrous metals are simply removed by utilizing magnets. The removal of non-ferrous metals (mostly aluminium, but also zinc, copper, lead) is based on the induction of eddy currents. An eddy current separator consists of a conveyor belt and a high speed, independent magnetic rotor, which is capable of generating a strong, very rapidly alternating magnetic field [25]. Once the conducting particles pass the rotor, eddy currents are induced and the non-ferrous particles are repelled by the magnetic field.

To sort out the non-rigid (foils, bags) from the rigid plastics, wind sifters are often applied. These devices blow or suck out the non-rigid plastic waste with one ventilator based on differences in specific weight (surface to mass ratio) [26]. Wind sifters are also able to separate paper contaminations (like etiquettes) from the rigid plastic fraction. Ballistic separators, which consist of a shaking screen, are as well capable of separating non-rigid and rigid plastics from each other.

A broad array of colours are contained in the plastic waste streams. Besides colour, plastic particles can vary in terms of opacity. Separation based on colour, using optical colour recognition sensors, can remarkably enhance the value of these recyclables. Likewise or light coloured, separated plastic particles can easily be recoloured, so they can match new design specifications. Sometimes, different plastic types can be separated based on their colours [27].

Contamination of one polymer in the matrix of another polymer material, usually results in a decrease of (mechanical) properties and reprocessing problems. A proper example of such a polymer pair that certainly must be separated is PET contaminated with PVC. At the processing temperature of PET, PVC will degrade and form the high corrosive hydrogen chloride gas. PET, on the contrary, will not melt at PVC processing parameters. For that reason, accurate and cost-effective sorting methods should be used and further developed. Automated separation methods can be subdivided into two categories, namely direct and indirect sorting techniques [20].
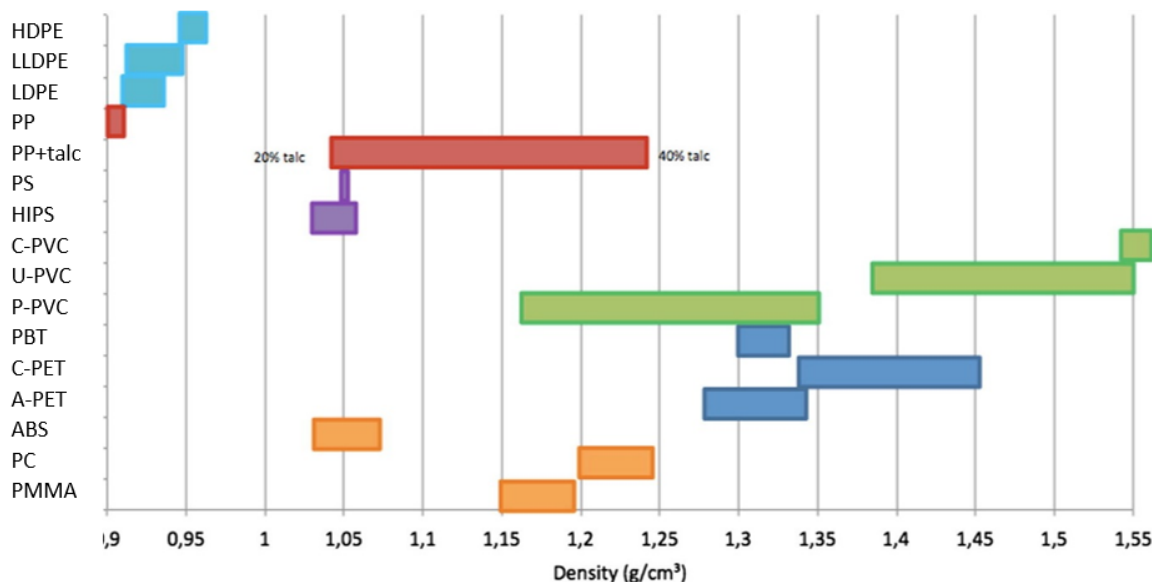

FIGURE 5. Density range of the most common polymers [15]. abbreviations: HDPE (high density polyethylene), LLDPE (linear low density polyethylene), PP (polypropylene), HIPS (high impact polystyrene), C-PVC (chlorinated pvc), U-PVC (unplasticized pvc), P-PVC (plasticized pvc), PBT (polybutylene terephthalate), C-PET (crystalline pet), A-PET (amorphous pet), ABS (acrylonitrile butadiene styrene), PC (polycarbonate), PMMA (polymethyl methacrylate)

Direct sorting methods are based on material properties like density and electrostatic characteristics. Most straightforward technique is the density-based sink-float method (Figure 6). By using water as flotation medium, polymers with densities below 1 g/cm³ (unfilled PP and PE) will float and are separated from the heavier polymers (PET, PS, PVC, ABS, ...), which will sink. To further separate the different polymers, density modifiers such as salts

particles are separated by different deflection in an electrostatic field.

Indirect separating techniques, on the other hand, make use of sensors capable of fast detecting and locating different types of polymers. FT-NIR (Fourier Transform Near Infrared) is by far the most applied technique for the sorting of plastic waste, as illustrated in Figure 7. However, this technique knows a few
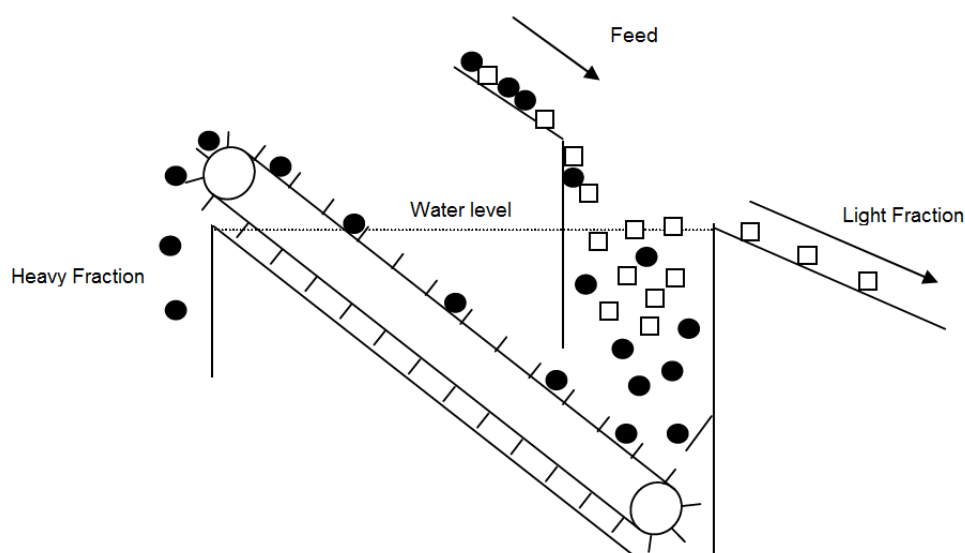

FIGURE 6. Principle of sink-float separation [26].

can be added to the water to create denser flotation media (heavy density separation). However, most polymer types have a density range, and these ranges often overlap as shown in Figure 5. For this reason, it is difficult to sort out polymers into mono-streams only based on density differences. To enhance the performance of density separation, density media separators like centrifuges and hydrocyclones can be applied. The machines improve the material wettability and also sort out plastic particles based on size and shape [20].

To separate polymer types with similar density ranges, more advanced techniques are needed. Selective flotation or froth flotation is based on the differences of hydrophobicity of polymer surfaces [28]. Through conditioning with wetting agents or physical treatments (flame, corona) the wettability of polymers can be improved. Air bubbles will attach to hydrophobic surfaces and will therefore induce flotation of the specific material. On the contrary, hydrophilic particles will remain completely wetted and stay in the liquid medium. Another promising technique is triboelectric separation, which is grounded on the surface charge transfer phenomenon [20]. Plastic particles are rubbed against each other and become oppositely charged. Subsequently, the

limitations; false readings are not excluded because of possible contaminations (paper, dirt) and this method has detections issues with black, dark or multilayer materials [15]. Besides FT-NIR, X-Ray detection (separation PVC containers) and laser sorting are upcoming sensor-based techniques for fast automated sorting of mixed plastic waste [7].
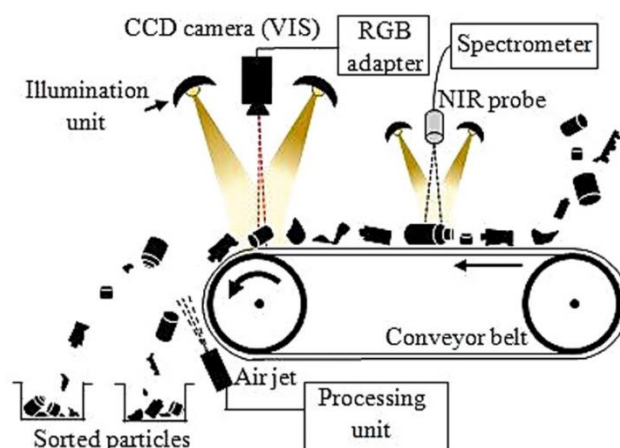

FIGURE 7. Principle of a spectroscopic NIR sorting technique [29].

4 CHALLENGES FOR MECHANICAL RECYCLING

When recycling mono- and mixed plastic products, different issues and challenges will occur [15], [20]. During the reprocessing of polymers, thermo-mechanical degradation can occur, as a combination of heat and mechanical shear. The other types of degradation can occur during lifetime, such as the exposure of plastic products to heat, oxygen, light or moisture [15]. Furthermore, the heterogeneous nature of the plastic waste makes it difficult due to immiscibility of the polymers and separation of the phases. The third main limitation is contamination of the mixture by different additives, fillers or even other polymers that are hard to recycle. During the reprocessing of these mixtures, often the processing temperature is set at temperature of the highest melting component, which can lead to overheating and degradation of polymers with lower melting point, and affect the final properties of recycled product.

4.1 Polymer incompatibility

The mechanical recycling of mixed polymers will inevitably lead to the formation of polymer blends [15]. The most common polymers are essentially immiscible in the melt phase. This can cause low mechanical properties of the final products, due to the low adhesion and phase separation between the polymer phases.

The simplest way to represent the miscibility of polymer blends is by using the following equation of free Gibbs energy [30], where the Gibbs free energy of the polymer mixture ΔG_{AB} has to be lower than the summation of the Gibbs free energy of the different polymeric constituents A and B:

$$\Delta G_{mix} = \Delta G_{AB} - (G_A + G_B) \leq 0 \tag{1}$$

$$\Delta G_{mix} = \Delta H_{mix} - T\Delta S_{mix} \leq 0 \tag{2}$$

Equation 2 consists of two terms, concerning the enthalpy of mixing (ΔH_{mix}) and the entropy of mixing (ΔS_{mix}). Blends can be miscible, partially miscible or immiscible depending on these thermodynamics. A polymer blend will be miscible and homogeneous if the value of the free energy of mixing is negative ($\Delta G_{mix} < 0$) and if the criterion for Equation 3 is positive [30]:

$$\frac{\partial^2 \Delta G_{mix}}{\partial \varphi^2} > 0 \tag{3}$$

Where ϕ is the volume fraction of polymer B. When the Gibbs free energy has a positive value ($\Delta G_{mix} > 0$), the blend will be immiscible.

The basic theory for the calculation of the Gibbs free energy was done by Flory and Huggins[31]:

$$\Delta S_{mix} = -k_B N(\varphi_A \ln(\varphi_A) + \varphi_B \ln(\varphi_B)) \tag{4}$$

$$\Delta H_{mix} = k_B T N \chi_{AB} \varphi_A \varphi_{AB} \tag{5}$$

$$N = n_A + n_B \tag{6}$$

The free Gibbs energy of mixing can then be given by the following equation:

$$\Delta G_{mix} = k_B T N \left(\chi_{AB} \varphi_A \varphi_B + \varphi_A \ln(\varphi_A) + \varphi_B \ln(\varphi_B) \right) \tag{7}$$

where k_B is the Boltzmann constant, T is the temperature, N is the total number of mole, φ_A and φ_B are the volume fraction of component A and B, and χ_{AB} the Flory-Huggins parameter which has to be negative that the spontaneous mixing happen.

The dimensionless Flory-Huggins parameter χ_{AB} is considered as a measure of interaction energy between the polymer A and polymer B in the blend. In reality however, this parameter is strongly dependent on the temperature, pressure and concentration. In that case, χ_{AB} can be calculated by making use of the solubility parameter δ, using the following equation:

$$\chi_{AB} = V_m R T (\delta_A + \delta_B)^2 \tag{8}$$

Where V_m is the mixing volume, R the gas constant, T the temperature and δ_A and δ_B the respective solubility parameters of polymer A and polymer B. The solubility parameter concept can allow a predictive capability of assessing the potential of miscibility of liquids. The solubility of a polymers is considered as the possible interactions between two polymers, divided into dispersive or Van der Waals forces (δ_{id}), polar forces (δ_{ip}) and hydrogen bonding (δ_{ih}). Hence, the solubility parameter can be written as [32]:

$$\chi_{AB} = \delta_{id2} + \delta_{ip2} + \delta_{ih2} \qquad (9)$$

Completely compatible mixtures are called homogeneous blends and show a one-phase morphology. On the other hand, immiscible blends can have different kinds of morphologies [40]: spherical particles, cylinders, fibres, sheets or co-continuous phases, as shown in Figure 8.
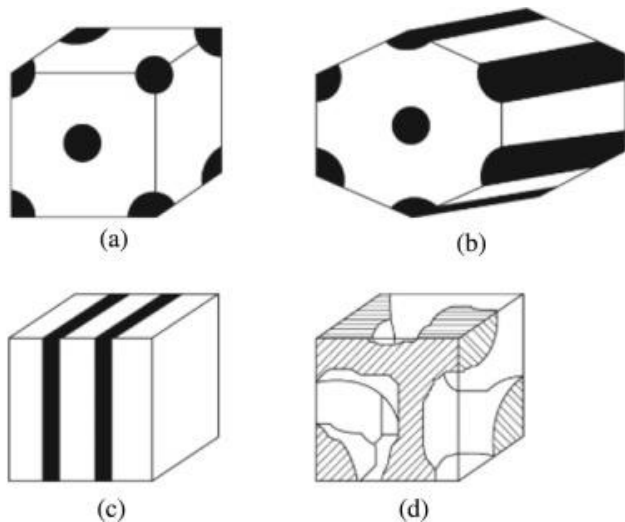

FIGURE 8. Morphologies of immiscible polymer blends: (a) droplets (b) cylinder (c) laminar and (d) co-continuous [15]

It is known that immiscible blends causing distinct phase morphologies have inferior mechanical properties compared with the virgin polymers. Additionally, coalescence of the dispersed phase might occur in an immiscible blend [33]. The coalescence phenomenon is considered as the merging of two or more particles into one new larger "daughter" particle. Such morphologies can strongly affect the final product performance, as properties such as impact strength and elongation at break are very sensitive to the dispersion and distribution of the second phase [33].

In a recent study, Hubo et al. [23] have compared mechanical properties of different recycled mixed polyolefin (MPO) blends. For mixtures based on recycled polypropylene (PP) and polyethylene (PE), they obtained low impact strengths, due to incompatibility of PP and PE in the melt phase, which consequently leads to the phase separation. Further, they have noted large variations in the values for tensile modulus, explaining that the impurities such as wood or non-PO in the mixtures will have a pronounced weakening effect during the tensile loading, while during bending test this effect was less pronounced. The phase separation that occurs during melting leads to an inferior transfer of stresses and strains within the material, resulting in lower modulus for the blends [34].

4.2 Polymer degradation

Environmental factors can cause changes in polymer, physical or chemical nature, resulting in bond scission and subsequent chemical transformations. Those transformations in polymers structure are categorized as a polymer degradation. It reflects in changes of material properties such as mechanical, electrical characteristics, in cracking, crazing, discoloration, phase separation or delamination[35]. There are several types of degradation: photo-degradation, thermal-mechanical or biological degradation.

Photo-degradation is related to the sensitivity of polymers to absorb the harmful part of the tropospheric solar radiation. Most of the polymers tend to absorb high levels of energy radiation in the UV part of the spectrum, which causes the activation of their electrons to higher reactivity and oxidation, cleavage, and other degradation types[35].

Thermal-mechanical degradation can occur during the thermal reprocessing of plastic waste as a result of overheating, both in terms of time and temperature[36]. There is a series of chemical reactions involved in thermal degradation which lead to physical and optical changes of properties, compared to the initially polymer properties[35]. During reprocessing, chain scission can occur, as well as chain branching. Depending on the initial molecular weight (Mw) of the polymer, one or the other mechanism will initiate, which will cause a reduction in Mw or an increase in Mw, respectively[15]. These mechanisms are shown in Figure 9.
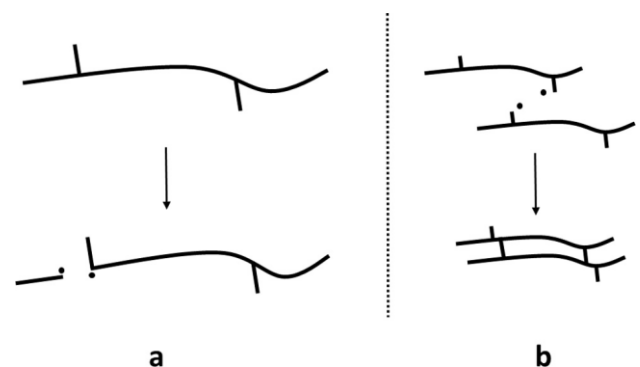

FIGURE 9. Random chain scission (a) and crosslinking (b)[15]

Thermal-mechanical degradation in most of the cases begins with a homolytic scission of a C-C covalent bond in the polymer backbone, causing generation of free radicals. These free radicals may undergo some chemical reactions such as disproportionation, causing chain scission, or crosslinking also known as branching[37].

Both degradation and immiscibility will strongly influence the rheological behaviour of the melt during processing. The rheological behaviour of polymer blends and recycled materials is often quite complex. The important factors are the chemical nature of the polymers in the mixtures, processing temperature, viscosity ratio, concentrations and processing [38] conditions [48].

5 ADDITIVES IN/FOR RECYCLED POLYMERS

Additives are universally incorporated in plastics. Finding plastics without additives is very unlikely, even more for their recycled counterpart. End-of-life plastics have a certain amount of degradation whereof the degree of deterioration of the properties depends on the lifetime of the plastics. To overcome the effect of degradation, or rather improve the mechanical properties of the recycled plastic (blends), additives are added to the recycled waste streams. First the remaining additives in the plastics such as: fire retarders, anti-oxidants and thermal stabilizers are discussed. Other additives which are mixed with the recycled polymers during reprocessing are: impact modifiers, compatibilisers, coupling-agents, glass and wood fibres, talc, chain extenders, etc. This incomplete list can be extended by many other additives and depends on the final requirements of the end-product and its applications.

5.1 Anti-oxidants, UV-and thermal stabilizers and fire retardant fillers

The properties of the recycled plastics are rarely the same as those of its virgin counterpart. Typically, they will be worse. Even polymers with short lifetimes are susceptible to degradation. This is mostly related to the influence of UV-light and/or oxidation [39]–[41]. During the first processing step, anti-oxidants are intermixed to prevent thermal oxidation during processing. Some plastics (such as PP with its tertiary carbon atoms and PET with its chromophoric groups) are also prone to photo-oxidation caused by a combination of UV-light and oxygen [42], [43]. Consequently, (re)processing polymers requires extra stabilisation. Common used stabilizers are hindered amines light stabilizers (HALS), which are anti-oxidants used as light scavengers. Other UV-stabilizers are nickel complex and carbon black. As for thermal oxidation, this is countered by hindered phenols (primary stabilizer) as radical scavengers. They provide longer product stability and preserve the melt viscosity.

Synergistically, phosphites and sulphites are used to counter the formation of hydroperoxides. They are also called secondary stabilizers and are used to stabilize during processing conditions [44]–[46]. After reprocessing, some of these additives can still be active/functional to stabilize the recycled end-product. As a means of determining the remaining amount of active thermal anti-oxidants, the oxidation induction time (OIT) can be used [39], [47].

Some polymers, like PVC, tend to degrade during processing solemnly due to thermal (and mechanical) influences. The degradation consists of dehydrochlorination and discolouration. Hence thermal stabilizers will be added, which react with the released hydrogen radicals to prevent further degradation of the processed polymer [48], [49].

Other waste streams, such of construction materials, transportation ELV, WEEE can contain fire retardants for safety reasons. However, in the past few years the regulations towards fire retarders has been altered. Older waste products mostly contain brominated or chlorinated flame retardants such as polybrominated biphenyls and polybrominated diphenyl ethers which can lead to the release of dioxins [50]–[52]. The use of these fire retarders are prohibited nowadays as they have been specified as hazardous. Although as it is established by REACH, these former approved fire retarders are "unintended impurities present in the final product and can be present in the starting materials or is a result of reactions during the lifetime or during the production process". It is advisable to separate that waste stream since they are not suitable for mechanical recycling. By X-ray fluorescence (XRF), the contaminated WEEE-products can be separated from the non-hazardous waste products [52]–[54]. The big variety of fire retarders on the market does not improve the recyclability of the waste. Very recently, the recyclability by reprocessing of different flame retardants in SPW was reviewed by Delva et al. [55]. Some examples of fire retardant fillers are: phosphorus, silicon, boron, nitrogen containing flame retardants and miscellaneous elements. Which fire retardant filler is used is strongly dependant on the polymer system, their mode of action, combustibility of the polymers, smoke release or toxic fume production. Often a mix of synergistic flame retarders is used to anticipate different types of combusting polymers. The molecules can influence the reprocessing in terms of rheology [50], [51].

5.2 Impact modifiers and chain extenders

Recycled post-consumer waste streams have one common trend: all of them show lower elongation at break and reduced impact resistance. By adding an elastomer to the recycled polymer, the impact resistance can be increased by preventing major crazing and ending the propagation of cracks, due to the dissipation of stress and the excellent elasticity of elastomers [56]–[58]. Studies for toughness improvement usually concern [59]–[68]:

➤ The content, shape, size and distribution of the elastomer filler

➤ The molecular weight of the components

➤ The interfacial adhesion with its interparticle distance and its effects on the mechanical properties in relation to the structure of the blend

➤ The influences of processing

➤ The stress-strain rate/temperature conditions.

To reduce the ecological fingerprint even more, the elastomers to optimize the recycled polymers' properties, can be recycled polymers themselves. Some examples are: poly(vinyl butyral) (PVB) from wind shields for polyamide (PA) toughening or EPDM from shoe soles or roofing applications, styrene butadiene rubber (SBR) and natural rubber (NR)/SBR scraps from automotive tires for PP [68]–[70]. However, the addition of impact modifiers have also a counter side: the rubber toughening of polymers usually results in a severe reduction in the tensile modulus and tensile strength [71]–[76]. Figure 10 shows, 20 wt% of the impact modifier SEBS-g-MA improved the impact resistance by almost a double with the trade-off de decreasing flexural modulus by only a fourth.

5.3 Compatibilisers

The great challenge of mechanical recycling is finding the cheapest method and at the same time the optimal separation technique. Post-consumer pure mono-stream polymer wastes are rarely found. As previously discussed, a solution to a 100% purified waste stream of commingled plastics has not been found yet. As many plastic waste stream consists of one or more impurities, the mechanical properties (originated from the low miscibility) will be feeble. Nevertheless most thermoplastics may be reprocessed with small amounts of impurities (< 5%) without excessive deterioration of mechanical properties. Except for the impact properties, which may only be improved
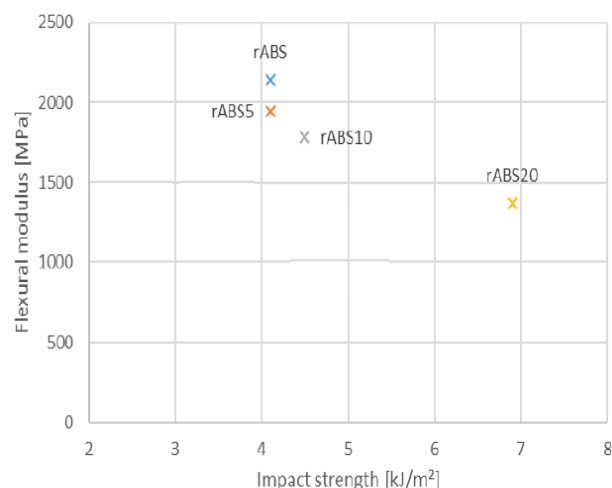

FIGURE 10. Increase of notched impact strength combined with a decreasing flexural modulus for recycled abs from weee and elv with (bleu x) no impact modifier, (orange x) 5 wt%, (grey x) 10 wt% and (yellow x) 20 wt% impact modifier sebs-g-ma (altered figure with permission from [77])

for mixed waste streams by adding an appropriate modifier, often refer to as compatibilisers [39], [78]–[81]. Compatibilisers can mainly be divided into three groups (Figure 11):

➤ Block copolymers
➤ Copolymers with functional groups
➤ Graft copolymers

They all attempt to minimize the interfacial distance and interconnect both phases by either using the same polymers as the matrix and the dispersed phase polymer or by interaction or reaction between the compatibilisers' moieties and the disperse phase and the compatible backbone polymers with the matrix. Grafted compatibilisers exist out of a grafted monomers and a backbone polymer. The latter can be preferentially the matrix polymer or an elastomer, the graft monomers are often maleic anhydride, glycidyl methacrylate, methacrylic acid, etc. [72], [74], [79], [80], [82]–[84].

The effectiveness of the compatibiliser strongly depends on: architecture of the block and/or copolymers, the reactivity of the moieties (when used) and the diffusion possibility of the compatibiliser across the interface. Condensation polymers offer the grand advantage of end-groups which have the possibility to react with the functional groups of the compatibiliser. Most important is that the interfacial tension of the compatibiliser is in between the one of both dispersed and matrix phase wherefore it will preferentially move towards the interphase. There are plenty of comparisons in literature of different types of blends, with the optimal amount and which compatibiliser one should select. However, they often lack a cost/benefit analysis and there are still no final guidelines on the best approach since every blend is different
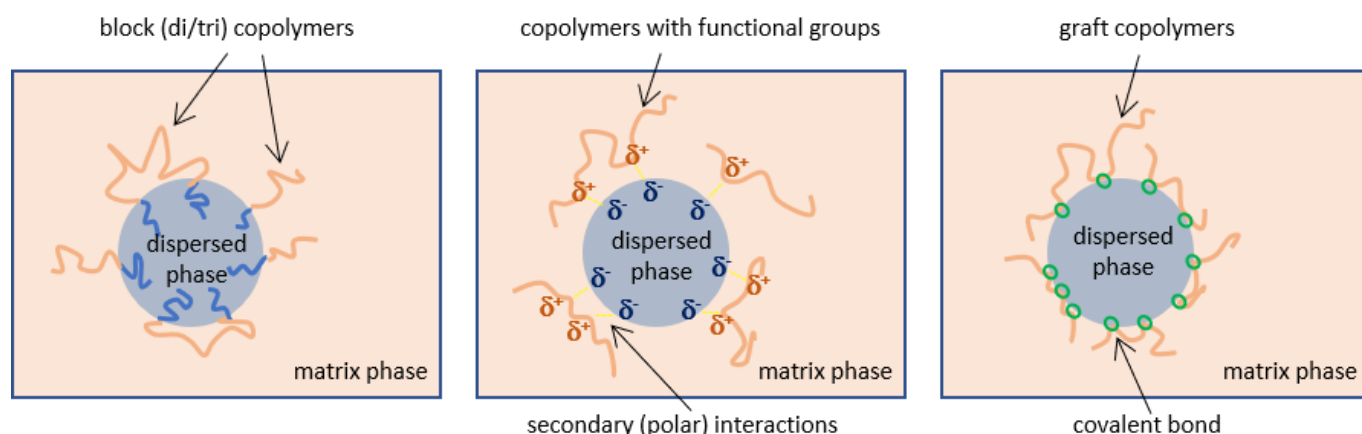
[62], [70], [73], [75], [81], [84]–[90]. A few examples of compatibilizing strategies available for post-consumer packaging waste is presented later in this manuscript.

5.4 Talc, glass and wood fibres

Just like virgin polymers, recycled waste streams can be filled with cheap yet effective particles, to improve their stiffness. This can be done by adding talc, glass or wood fibres, nanoclays or other micro- or nanosize particles which should have higher stiffness than the matrix polymer. The fillers of commingled plastics from their previous lifetime might cause difficult processing and undesirable changes in mechanical properties in the other components. This will eventually lead to a deterioration of the entire product. PA and PP are often mineral filled or glass fibre reinforced in the automotive industry to ensure the parts will endure high strength at elevated temperatures [70], [91]–[93]. As for most of the fillers, they are not compatible with the matrix. A suitable modifier (coupling agent) should be selected in order to improve interfacial adhesion. This modifier preferentially has polar groups in its structure, due to the polar groups or ions present in the fillers. The addition of the modifier will consequently change the morphology of the blend to a better dispersion and usually also better processing conditions in terms of a more continuous flow. As for the optimal amount of reinforcing filler, which is intermixed during (re)processing, this depends on its final application of the product and typically amounts between 20% and 60% [68], [94]–[96].

6 Examples of mechanical recycling

6.1 Post-consumer PET (bottle-to-bottle) recycling

Poly(ethylene terephthalate) (PET) is a semi-crystalline thermoplastic condensation polyester [97]. The PET demand accounts for 7,1% of the total European plastic demand corresponding to a total demand of 3,5 MT (PET fibers not included) [3]. It is mainly used in packaging applications such as bottles, containers, trays and foils. Good mechanical strength, low permeability towards moisture and oxygen, inertness, low specific weight and high clarity are some of the key properties which contribute to the applicability of PET in packaging applications [98]. Next to the packaging applications, a huge end-market for PET is the use as fibers in textile industry (approximately 1 MT in 2011 in Europe [99]).

Petcore, the European PET association, estimated the overall collection rate in Europe for 2014 for PET bottles and containers at 57% by comparing the collected waste with the PET placed in the market [100]. The overall collection rate is often confused with the overall recycling rate, which is off course more valuable for the European policy but also more difficult to calculate [101].

As previously mentioned, separation post-consumer waste in their mono-polymers is essential to reach high-quality recycling. PET bottles can quite easily be separated from other polymers using automated or hand sorting techniques. It should definitely be separated from PVC, as previously explained, degradation products of PVC are known to facilitate PET degradation by

catalyzing chain scission reactions during reprocessing [102]. Normally the PVC content should be lower than 0.25% [100]. It is typically separated (depending on the country) in clear, blue and green PET.

Different recycling processes are industrially being used to mechanically recycle the sorted post-consumer PET bottles. Although these recycling systems all are very specific, typically the following process steps are found [103], the order of which can vary:

➤ Conventional preparation (grinding, washing, removal of caps and labels)
➤ Re-extrusion (melt reprocessing)
➤ Solid-state polycondensation (removal of various contaminants and/or increasing of intrinsic viscosity)

The first conventional treatment steps consist of grinding, washing and removing caps and labels from the PET bottles. The washing aims at removing all surface contaminants and is usually done in a NaOH water solution. Remaining polyolefin contaminations can also be removed in this step by float-sink separation.

The re-extrusion step is done by extruding the flakes at temperatures above the melting temperature of the PET (>260 °C), mostly combined with the use of vacuum to remove low-molecular contaminants.

Solid-state polycondensation (SSP) is typically performed to restore the intrinsic viscosity of post-consumer PET flakes. Typically post-consumer PET displays lower intrinsic viscosities (due to reduction in molecular weight, presence of moisture, hydrolysis, etc.) compared with virgin PET grades. SSP involves heating of the PET at a temperature between the glass transition temperature and the melting temperature in a reactor. Condensation reactions occur between the chains terminal groups in the amorphous phase of the polymer, in a temperature range of 180–240 °C. The reaction proceeds under vacuum to remove by-products. Additionally, at these elevated temperatures, post-consumer contaminants from inside the flakes will be able to diffuse to the surface to be removed by vacuum-extraction (if residence time is sufficient).

Alternative processes to increase intrinsic viscosity is by adding chain extenders (during solid-state polycondensation or during reactive extrusion) which can act as cross linkers between the individual PET chains leading to increased molecular weight and intrinsic viscosity [104].

An example of an industrially used recycling process is the "super-clean PET recycling process based on pellets". A scheme of this process is shown in Figure 12. This process combines all of the above mentioned process steps.
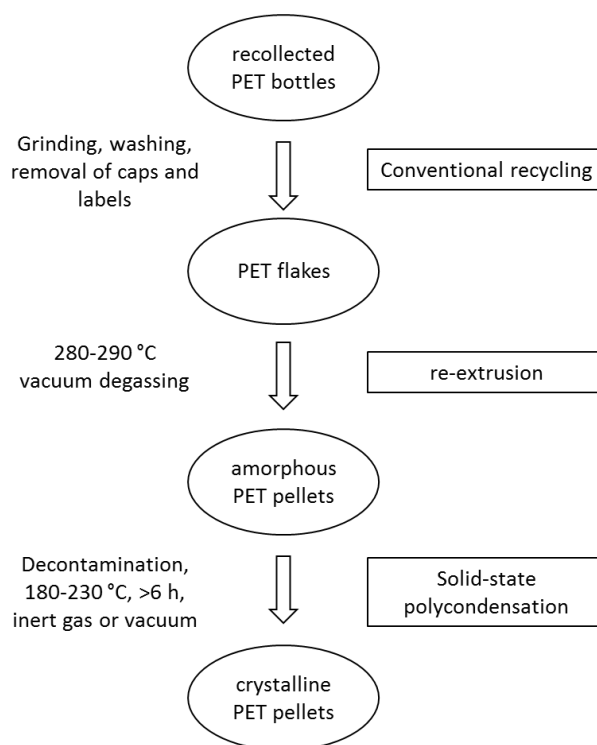

FIGURE 12. Scheme of pet super-clean recycling process based on pellets. adapted from [103].

Typically, the reclaimed PET pellets (mostly diluted with virgin PET - depending on cost and properties) can then be used to produce new bottles, containers, fibers or thermoforms. Nowadays, up to 50% of a PET bottle can be made from post-consumer PET bottles [103]. In thermoform applications on the other hand this percentage can easily be higher.

rPET is often re-used in new food contact applications, however, for safety considerations, each recycling process has to follow a safety assessment procedure governed by the European Food and Safety Agency (EFSA). The evaluation principle of EFSA is based on the cleaning efficiency of the recycling process. This efficiency is determined by so-called challenge tests, where PET is deliberately (over)contaminated with (model) contaminants [105]. These evaluations are published on-line, e.g. the EFSA evaluation of the EREMA process, EREMA is a machine manufacturer for plastic recyclers [106].

Nowadays, clear and light-colored (mainly blue and green) PET bottles are easily collected and separated for secondary recycling in Europe. However, opaque bottles and thermoforms are currently not recycled. Several countries in Europe are currently evaluating the possibility to additionally recycle PET thermoforms and other difficult to recycle PET waste streams (e.g. in France

[107] and in Belgium [108]). The main difficulties until today include:

➤ Complex compositions (multilayers, inks, glues, absorbent pads, etc.)

➤ High rPET content in thermoforms

➤ PET flakes with lower intrinsic viscosity

➤ Equipment of recycling plants (more fine dusts, brittleness of thermoforms, etc.)

➤ Food contact regulations

6.2 SPW from post-consumer packaging waste

The diverse applications for plastic packaging materials lead to a large variety of compositions of packaging which oftentimes consists of multiple layers in order to benefit from the different properties each polymer brings to the table. These packaging materials generally have a short lifespan and end up fairly soon and abundantly in the waste. Plastic packaging products are mainly produced in LDPE, PP, PET, HDPE, PVC and PS [3], so these polymers will also dominate the composition of the post-consumer plastic packaging waste.

The collection systems for plastic packaging waste in Europe differ from country to country. Countries like Germany and the Netherlands use collection systems which allow all plastic packaging waste to be collected together. Belgium has a similar system which collects drinking cartons, metal packaging and plastic bottles, i.e. the so called PMD-system. In order to anticipate on the Eropean Circular Economy Action plan (EU Commission, closing the loop, [8]), an extension of the Belgian PMD system is taking place in a phased manner. In short, this entails the extention of the plastic fraction of the PMD allowing other plastic packaging materials such as foils [109].

This extension of the plastic waste stream implies a more complex waste stream as the composition changes substantially. These changes in composition in turn demand other and better sorting and separation processes to ensure a pure plastic waste end stream for recycling.

The PET fraction mainly originates from PET bottles and thermoformed trays. The bottles are initially (either by consumer or recycling plant) separated from the other products and are send to the bottle-to-bottle recycling plants (see above). The other fractions eventually end up in sink-float sedimentation tanks to produce a light fraction (i.e. float) and a heavy fraction (i.e. sink) [110]. The light fraction mainly consists of polyolefins (PE and PP) and is therefore used to produce different qualities of mixed polyolefin (MPO) streams, either 'hard' MPO's (rigid packaging) or 'soft' MPO's (flexible packaging) [111]. The heavy sink fraction is currently not being recycled and is prepared (e.g. lowering of the chlorine content) and send to energy recovery.

Despite the fact that the PE and PP in these MPO's are quite similar (i.e. they all consist of carbon and hydrogen atoms), even these mixtures are often not miscible due to differences in molecular structure and form heterogeneous blends which in turn have quite low properties compared to virgin polymers due to the formation of weak interfaces [23]. Therefore, these MPO waste streams are nowadays mechanically recycled mainly into 'low-quality' products such as garden furniture, outdoor flooring and traffic signalization elements by industry [112].

As depicted before, different strategies can be used to upgrade the properties of mixed polymers. Likewise for MPO's the addition of stabilizers, compatibilisers and/or impact modifiers can be required to level the properties compared to virgin grades. Typically copolymers (e.g. ethylene propylene diene monomer (EPDM) [113], ethylene-propylene rubber (EPR) [114] or block copolymers (e.g. SEBS [115]) are used to compatibilise polyolefin blends. Recently, Eagon et al. [116] have very carefully synthesized well-defined di-block and tetra block copolymers which increased the toughness of PE/iPP blends significantly even at very low percentages (1 wt%) as can be seen in Figure 13 (a). They point out the importance of molecular weight, which must ensure an increase in number of entanglements between the blocks and the homopolymer chains in the melt state. A sketch of the interface between the PP and PE is shown in Figure 13 (b).
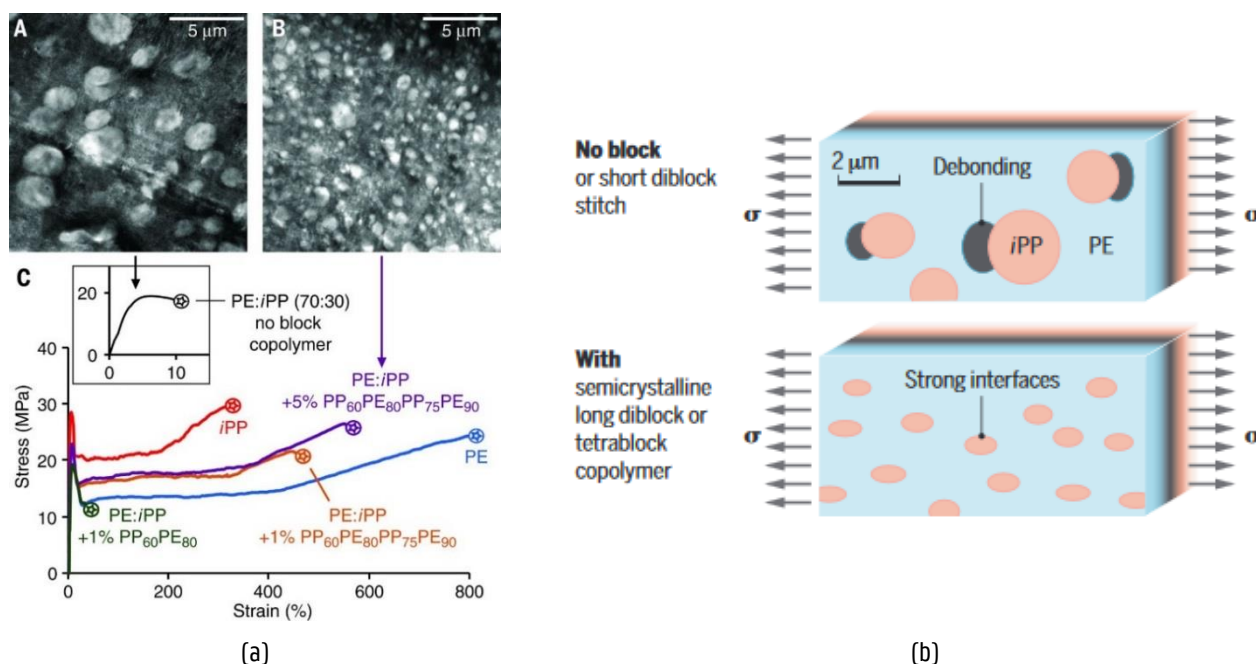
FIGURE 13. Tensile properties of PE/iPP blends with and without addition of long di-block or tetra-block copolymer [116] and (b) Schematic interpretation of interfacial strength between iPP and PE with and without addition of long di-block or tetra-block copolymer [117]

6.3 SPW from WEEE

The production of electrical and electronic equipment or (EEE) is growing at an increasing pace due to our rapid economic growth and growing demand for consumer goods [118]. This vast production rate is accompanied by a large amount of waste electronic and electrical equipment (WEEE). This sector, next to the transportation and appliances area, is expected to grow even more. The rapid development of the EEE technology combined with the lower life cycle of most present-day products poses a significant issue as far as their disposal is concerned. In view of this environmental problem concerning the management of these WEEE a legislation was drafted to improve reuse, recovery and recycling of the latter. Thanks to the European Directive (2000/53/EC) and the WEEE directive (2002/96/EC) at least 70-80% of materials of end-of-life vehicles (ELV) and WEEE have to be recovered in the form of energy and/or materials [119]. This means that a large amount of polymers reenters the market for reprocessing. A study by Achilias et al. [119] demonstrates that a typical WEEE fraction contains 20-30% plastics. The general composition of the plastic fraction itself is depicted in Figure 14. As can be seen from this graph the main constituents of the WEEE plastic fraction are ABS, high impact polystyrene (HIPS), polycarbonate (PC), PC/ABS and PP.

To better understand and anticipate the performance of these recycled products further investigation regarding the changes in properties during recycling still has to be done. The main problem with these materials is the variability in the product composition due to the presence of polymer mixtures, additives or contaminations. In addition, it is well known that polymers are subject to degradation, both by daily use as during processing of the materials [120]. In its everyday use, polymers are susceptible to photo-degradation, oxidation, leaching of various additives (e.g. plasticizers) and other weathering phenomena. These effects are furthermore exalted during melt
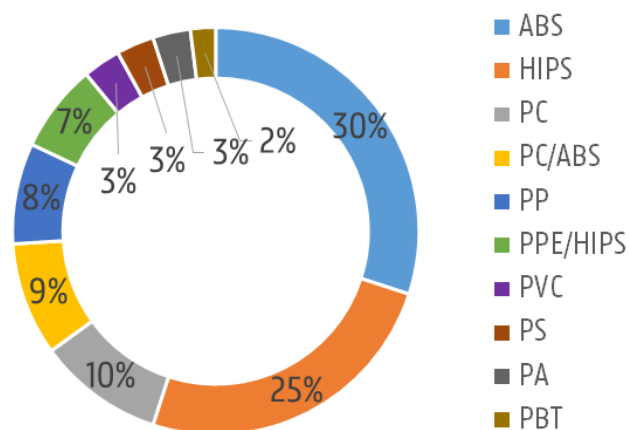

FIGURE 14. Typical WEEE composition according to Achilias et al. [119]

reprocessing operations causing the properties of the recycled materials usually to be worse than that of the virgin material. As EEE products typically have a long lifespan compared to other polymer applications (e.g. packaging), degradation will play a more prominent role in influencing the final properties of the recycled material. Here, during its lifetime, stabilizers are slowely depleted/degraded until oxidation and degradation of the polymer chains can occure reducing the quality of the recycled product.

Possible ways of upgrading these recycled plastics include blending them in small portions with their virgin grade [121]. Often however, depending on their lifetime and amount of weathering the plastic has undergone, these blends suffer from more or less limited miscibility requiring the use of suitable compatibilisers for stabilization of the system [121], [122].

Another way of improving the polymer properties is by blending the recyclates with suitable additives. Pospíšil et al. [122] made an extensive overview on the stabilization of recycled plastics by upgrading them in a melt blending process with suitable additives. They stress the importance of the history of the plastic as the application determines the amount of weathering the material has undergone. Structural changes take place by reprocessing, heat ageing or weathering that differentiate the WEEE plastic from its original virgin form. The gravity of the changes is dependent on the service time of the plastic and the aggressiveness of the environment. Careful selection and analysis of the plastic waste is incumbent for reuse into new applications.

Frequently stabilizers are re-added to the system because they have been consumed during their lifetime and processing. In this case heat stabilizers such as hindered phenols or hindered amines are usually combined with phosphorous or sulfuric type secondary stabilizers [44], [46], [123]. UV-stabilizers prolong the lifetime of the plastics by absorbing harmful radiation and deactivate the latter by radiationless processes [124]. Metals, fillers and pigments can be added to the recyclate as well, yet one should be careful of residual components that could catalyze the decomposition of peroxides forming radicals that accelerate degradation. In this case metal ion deactivators, antacids and deactivators for residual fillers can be added.

7 Conclusions

This paper has reviewed the current state-of-the-art in the field of mechanical recycling of SPW. Mechanical recycling is the current industrially ubiquitous technique for the recovery of waste polymers. Different technological aspects such as collection and sorting have been discussed, as well as the materials science behind the main challenges associated with an efficient mechanical recycling economy such as contamination, degradation and the mixing of different plastics types in waste. Furthermore, several industrial technologies were highlighted focusing on post-consumer PET bottle-to-bottle recycling, SPW from post-consumer packaging waste and SPW from waste from WEEE. In addition, various additives commonly found in or added to recycled polymers were discussed.

Ready to learn more?

This introductory review has been broad, rather than deep. If you would like to get acquainted in more detail with our work, you could check our publications (https://biblio.ugent.be/person/802000178474) or have a look at our website (www.cpmt.eu).

Some of our key publications:

➤ Waste Management: Ragaert, Kim; Delva, Laurens; Van Geem, Kevin. **Mechanical and chemical recycling of solid plastic waste**

➤ Waste Management: Marieke T. Brouwer , Eggo U. Thoden van Velzen, Antje Augustinus , Han Soethoudt,Steven De Meester, Kim Ragaert. **Predictive model for the Dutch post-consumer plastic packaging recycling system and implications for the circular economy**

➤ Waste Management: Delva, Hubo, Cardon, Ragaert: **On the role of flame retardants in mechanical recycling of solid plastic waste**

➤ Journal of Cleaner Production: Sophie Huysveld, Sara Hubo; Kim Ragaert; Jo Dewulf. **Advancing circular economy benefit indicators and application on open-loop recycling of mixed and contaminated plastic waste fractions**

➤ Polymer Engineering and Science: Ragaert, Kim; Hubo, Sara; Delva, Laurens; Veelaert, Lore; Dubois, Els. **Upcycling of Contaminated Post-Industrial Polypropylene Waste: a Design from Recycling Case Study**

➤ Polymer Engineering and Science: Peelman N., Ragaert P., Ragaert K., Erkoc M., Van brempt W., Faelens F., Devlieghere F., De Meulenaer B., Cardon L. **Heat resistance of biobased materials, evaluation and effect of processing techniques and additives**

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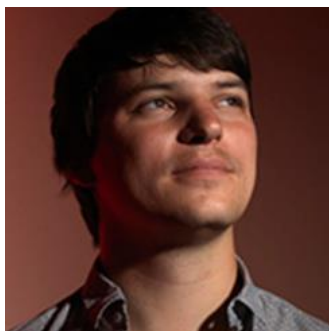
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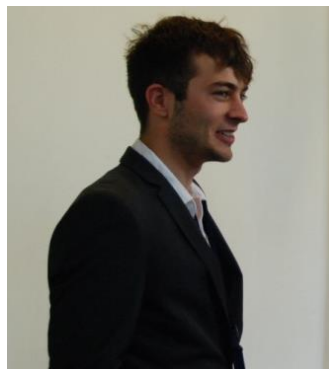
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