Chapter

Recent Advances in Pre-Treatment of Plastic Packaging Waste

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Abstract

There is an urgent need to close the loop of plastic waste. One of the main challenges towards plastic packaging waste recycling is the presence of a variety of contaminants. These contaminants include organic residues, additives, labels, inks and also other plastic types that can be present in the waste stream due to mis-sorting or in multimaterial structures (e.g. multilayer films in packaging). In this context, pre-treatment processes are a promising route to tackle the difficulties that are encountered in mechanical and chemical recycling due to these contaminants. This chapter gives better insight on the already existing pre-treatment techniques and on the advances that are being developed and/or optimized in order to achieve closed-loop recycling. Some of these advanced pre-treatments include chemical washing to remove inks (deinking), extraction methods to remove undesired plastic additives and dissolution-based pre-treatments, such as delamination and dissolution-precipitation techniques.

Keywords: Recycling, Plastic Packaging, Circular Materials Economy, Pre-treatment, Chemical cleaning

1. Introduction

The largest share of post-consumer plastic waste is plastic packaging, [1, 2] which is typically composed of among others low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), poly(ethylene terephthalate) (PET) and polystyrene (PS). [2] The European Commission has set recycling objectives to move towards a more sustainable plastic economy, namely: (i) achieve 55% recycling of plastic packaging by 2030, (ii) increase fourfold the sorting and recycling capacity (iii) and produce only reusable or recyclable plastic packaging. [3]

The main challenges faced during the recycling processes of plastic materials is the complexity of this waste stream. Plastic packaging waste streams typically consist of a mixture of polymers and contaminants, such as paper, organic residues, odor constituents, adhesives and inks containing halogens and metals. [4] The presence of additives that are incorporated in the plastics during the manufacturing process also bring complications during mechanical and chemical recycling. [1, 5] The most used additives in packaging materials are: plasticizers, antioxidants, acid scavengers, light and heat stabilizers, lubricants, pigments, antistatic agents, slip compounds and thermal stabilizers. [5] However, during mechanical recycling
these additives are blended in the recycled materials, which might be a potential health risk for consumers, especially in food contact materials. [4] For example, flame retardants such as persistent organic pollutants (POPs), phosphorous flame retardants and phthalates have been found in children toys. [5] Brominated flame retardants were also detected in food contact materials and household products. [5] Some additives can also have a direct impact on the recyclability of the plastics and even lead to the degradation of the plastics. This is the case for metal-containing additives, such as metals salts or oxides like Fe$_2$O$_3$, Cu$_x$O and ZnO, that will form pro-oxidants and photo-oxidation catalysts, promoting the degradation of the plastics during the reprocessing phases in mechanical recycling. [5] Furthermore, leaching to the environment of substances such as metals, volatile organic compounds, phthalates, polycyclic aromatic hydrocarbons, among others, is also a problem faced during different stages of mechanical recycling. [5] Another recycling issue is the presence of multilayer materials that typically consist of immiscible polymers (e.g. PET and polyethylene (PE)) that lead to recyclates with low mechanical properties. [6] Complications also arise during chemical recycling when e.g. halogenated compounds are released during the polymer decomposition, which can cause corrosion of the process equipment and reactors. [4, 7] Also for chemical recycling technologies, multilayer materials might cause complications as they typically contains polymers such as PET layers, which lead to the release of oxygenated compounds. [4, 6]

With the currently applied pre-treatment technologies, such as sorting, washing, float-sink and grinding, there are still contaminants present in the post-consumer plastic waste, [4] as these techniques are not able to thoroughly clean the polymers and remove the impurities embedded in the polymer structure. [8] A more circular economy for plastics would need more advanced pre-treatments such as chemical washing technologies, deodorization, deinking, delamination and solvent-based extraction methods. Deodorization and deinking techniques enable the removal of odor constituents and inks present in the plastic waste, respectively. Delamination can tackle the issue of multilayer materials by selective decomposition of polymer layers and/or adhesives. [6] Solvent-based extraction methods are able to remove several additives from the polymer matrix. Solvent-based extraction methods can be divided into two groups: dissolution-precipitation and solid–liquid extraction methods, which include ultrasonic extraction, supercritical fluid extraction, microwave-assisted extraction and accelerated solvent extraction.

2. Pre-treatments

The aim of this chapter is to provide a review of the advances in pre-treatment proposed in literature for the purification of plastic waste. Thereto, technologies that are applied from lab-scale until industrial scale are discussed. The first section of this chapter focusses on the more conventional pre-treatment steps comprising sorting and washing of plastic packaging. Subsequently, the recent advances in deodorization, deinking and delamination of plastic packaging will be discussed in depth. In the last section, a detailed overview will be given on solvent-based methods, comprising dissolution-precipitation and extraction technologies.

2.1 Conventional pre-treatment steps

In Europe, extended producer responsibility (EPR) is set up for the collection and recycling of packaging waste. Via EPR a producer’s responsibility for a product is extended to the post-consumer stage of a product’s life cycle. [9] This is generally
translated to an environmental fee that producers need to pay to have their products managed through the so-called producer responsibility organization (PRO). A PRO is a collective entity set up by producers or through legislation, which becomes responsible for meeting the recovery and recycling obligations of the individual producers. [10] Depending on the country, an individual or co-comingled kerbside collection system can be implemented, a deposit-refund system might be organized or combinations thereof. In Germany, for instance, there are two separate collection systems for plastic packaging, namely a deposit system for PET bottles and the Dual System, where packaging and non-packaging wastes made of plastic, paper, metals and composite material are disposed for commingled collection. [11]

Depending on the particular collection scheme, the waste stream will comprise a whole range of different packaging products and contaminants. [12] Due this compositional complexity, sorting of plastics in a Material Recovery Facility (MRF) is an essential step to provide high quality materials for the recycling industry. [13] An MRF comprises sorting units such as waste screening, air separation, ballistic separation, magnetic separation, eddy current separation, sensor-based sorting such as near infrared technology (NIR) and manual sorting in a certain configuration in order to generate bales as pure as possible. The output materials achieve purity levels up to more than 97%. [13] However, even after sorting, the generated plastic bales still contain certain substances, such as polymers, paper, and organic residue, present in levels between 10% and 20% of the total mass of the end-of-life packaging product. [4]

These bales are sent to recycling factories, currently mainly mechanical recyclers, that wash and regranulate the plastics. Figure 1 illustrates a typical process chain applied at a recycler’s plant.

The recycling process starts by reducing the size of the plastic products via a crude shredder. Henceforth, the shredded particles are transported via a conveyor belt to a first washing step. This is, for instance, a rotating drum washer where rocks, metals and glass are separated gravitationally; a water flow provides the washing. [1] This step is followed by a second washing by means of a friction washer. The plastic surfaces are here cleaned of organic residues, adhesives and glued-on labels that stick to the plastics by intensive mechanical agitation. The plastic material is transported via a screw, while dissolved impurities, fines, and process water are discharged through sieves. [15] The plastic particles are now fed into a miller to further reduce the particle size. From this point, they proceed to the float-sink installation, where a density separation of polymers is performed. Polymers with a density lower than 1 kg/L will float (e.g., PE and PP), whereas polymers with a density higher than 1 kg/L will sink (e.g., PET, PS and PVC). As
this is a water-based technique, the flakes simultaneously undergo an extra washing. [1] After the float-sink step, the flakes are dried via a mechanical and/or a thermal dryer. Finally the plastics are sent to extrusion, which includes a melt filtration to remove fractions such as wood, paper, aged rubber particles and higher-melting polymers (e.g. PET in PP processed at 220°C). [16]

Most recycling plants use water to wash the plastics, whether or not at elevated temperature. In some cases sodium hydroxide (NaOH) or a detergent can be added as washing additives in order to reduce surface contamination such as dirt, labels and glue. [17] Logically, a water-based washing step cannot achieve 100% efficient removal of the heterogeneous substances that are present on a post-consumer plastic waste stream, even not when caustic or detergents are added. Therefore, more advanced purification steps are under developing at lab or pilot-scale or are being integrated before, during or after the extrusion step in order to provide recyclates with a higher purity. However, even for more advanced pre-treatment steps sorting and cleaning with an initial water-based washing step to remove organics, papers, and polymers e.g., with a friction washer or density separation, is probably still a key step within the plastic recycling chain.

2.2 Deodorization technologies

One of the main hurdles towards high-end recycling of plastics is the presence of odorous constituents. Due to the persistent odor that remains after washing with specialized equipment such as friction washers, large volumes of plastic packaging waste are currently only suitable for downcycled applications, such as plant trays, compost bins, street or garden benches, etc. [18] In several scientific papers, the odor profile of different plastic packaging products is characterized. Especially the heterogeneity of odor components on packaging material in terms of physicochemical properties is described. This is illustrated by Figure 2, which gives an indication of the abundance of chemical groups that were detected on post-consumer waste plastics.

In total, over 400 volatile organic components (VOCs) are detected, which are here divided into 19 subcategories. With 60 and 56 components, respectively, esters and alkanes are two of the main functional groups found on plastics. However, their physicochemical properties are fundamentally different. This physicochemical heterogeneity of odor components makes efficient deodorization very challenging. Yet, most recycling plants only apply a water-based washing step, in best case with

![Figure 2](image)

Number of VOCs detected on plastic materials, divided into subcategories based on their chemical structure. Adopted from [19].
addition of a detergent. Logically, scientific and industrial experience have shown that the water-based washing media are insufficient to become recyclates with an acceptable odor threshold. Table 1 shows an overview of different scientific studies that investigated the efficiency of industrial recycling plants in term of deodorization of plastic waste.

As water-based treatments are insufficient, the demand for new and improved deodorization technologies is increasing. Although, industrial application of such technologies remains relatively limited to date. An interesting option is the use of a solvent-based approach in order to remove the more hydrophobic constituents. A commercial solvent-based process applies hot ethyl acetate to clean polyolefin-based packaging products. [23] Scientific research at lab-scale has indicated that with a batch-wise extraction using ethyl acetate at 65°C, an average removal of analyzed odor components of 90% is feasible. [8] Taking into account the benefit that can be made with a continuous counter-current process, polymers with relatively high purity levels can be achieved.

Another solvent that is tested to remove, among others, odorous constituents from a HDPE waste stream is poly(ethylene glycol) (PEG). PEG is considered as a relatively eco-friendly solvent and is getting attention due to its low volatility and toxicity compared to conventional solvents, and its higher miscibility with organic compounds compared to water. [24] A batch lab-scale extraction with PEG has shown that the quantity of VOCs was reduced with 74% after PEG extraction at 100°C. Hence, PEG is considered to be a promising solvent towards deodorization. [24] A schematic representation of a deodorization by means of PEG can be found in Figure 3a.

Applying solvent-based technologies might be quite expensive, taking into account the extra capital expenditures (CAPEX) and operating expenditures (OPEX) that are typically linked to such technologies. This is often not preferable in plastic recycling, given the associated typically low profit margins. [4] Applying chemicals such as solvents and detergents on a relatively highly contaminated waste stream, comprising next to odorous constituents also glues, paper, inks, additives, degradation products, non-target polymers, etc., should preferably be able to remove a large range or even all of such substances in order to be interesting from economic perspective. In this perspective, such technologies are not only investigated towards removal of odor components, but also towards removal of a broader range of impurities. The application and development of more solvent-based

| Material       | Treatment                                      | Result                                                                 | Ref. |
|----------------|-----------------------------------------------|-----------------------------------------------------------------------|------|
| LDPE bags      | Hot-water at lab-scale                        | Considerable odor intensity remained (6.3 out of 10)                  | [20] |
| MPO films      | Industrial recycling process                  | No reduction observed                                                 | [21] |
| MPO films      | Industrial recycling process                  | <65% reduction after washing increase with a factor 2–6 after extrusion compared to the washed films | [18] |
| PP rigid       | Industrial recycling process                  | Odor intensity ratings declined from 7.4 to 4.0                       | [22] |
| PE film        | Cold (25°C) and hot (65°C) water at lab-scale | 54% and 63% removal of VOCs                                           | [8]  |
|                | Cold (25°C) and hot (65°C) caustic + detergent at lab scale | 68% and 78% removal of VOCs                                           |      |

Table 1. Overview of scientific literature to evaluate odor removal from plastics achieved by going through a full-scale water-based industrial mechanical recycling facility and their respective main results.
technologies and their efficiency towards removal of different substances will be discussed more in detail in Section 2.5.

Besides applying extraction techniques, also the use of air to remove odorous constituents, as shown in Figure 3b. A commercial available example is the ReFresher technology with the INTAREMA® TVEplus® machine (EREMA Engineering Recycling Maschinen und Anlagen Ges.m.b.H., Ansfelden, Germany). This equipment applies heated air directly to the extruded pellets for flushing out volatile contaminants and simultaneously removes the air via a degassing unit. Applying a hot air stream during a few hours can significantly reduce the overall odor intensity of recycled HDPE pellets with an efficiency varying between 51.0 and 99.3%. [25] A disadvantage of this technique is the relatively long contact time that is needed to achieve the maximum feasible removal efficiencies, typically between 4 and 7 hours. Depending on its size, the ReFresher has a capacity between 350 kg/h and 4000 kg/h.

Likewise, steam can be applied to remove VOCs from plastic materials (see Figure 3c). A study has shown that an increased VOC reduction is achieved via steam stripping compared to hot air stripping. [24] Thereto, post-consumer HDPE was treated at a lab-scale distillation unit for 2 h. The produced vapor flowed through the plastic bed and left on the top, passing to a condenser. An overall reduction of volatile components above 70% was reported.

A similar approach is the use of a degassing system during extrusion is investigated towards the removal of VOCs. [19] Different methods are used e.g. degassing by vacuum, thermal degassing or degassing with the help of ultrasound. [26] A study to the removal of VOCs from plastics via a vacuum degassing system showed that the odor concentration was reduced with around 37% after three degassing steps. Devolatilization is considered to be a complex process as the correct choice of temperature and shear profiles, along with screw configuration and placement of venting influence the removal efficiencies of VOCs. For instance, a higher temperature and pressure during extrusion can increase the volatility of the moisture content and permit water and/or other volatile materials to be released. [27] Figure 4 shows a typical set-up for devolatilization of LDPE.

Another investigated method to minimize odor on post-consumer plastic packaging waste is through the use of a probiotic bacteria solution during the recycling process. In a recent study, a commercial probiotics formulation was investigated at a pilot scale before the washing step. [29] Significant differences in the overall odor intensity of the untreated reference sample and the sample treated with probiotic bacterial cultures were obtained by applying the probiotic bacterial cultures to the input material, followed by 40 days of storage. Especially those substances that most likely originated from microbial degradation of organic matter are reduced up to 70%. However, further improvement and investigation to industrial implementation of probiotic treatment is required.
A more established option to remove VOCs from plastic waste is the addition of high specific surface adsorbents during the extrusion process. [21] A great variety of adsorbents are available on the market. As they are added to a polymer in melting phase, VOCs can adsorb on the adsorbing agents’ surfaces. It is stated that adding 0.30 wt% of a certain adsorbents such as zeolite and activated silicate, can significantly reduce the amount of VOCs coming from post-consumer HDPE with approximately 50%. [30] A similar more recent development is the addition of reactive additives that undergoes a chemical reaction with the functional groups of odor-causing substances and, hence, convert them into non-volatile components. [31] Examples of such a commercial available additives are zinc ricinoleate (Tego Sorb PY 88; Evonik Industries AG, Essen, Germany) and Recycloblend 660 (PolyAd Services). However, scientific studies to quantify the effectiveness of such additives are scarce.

2.3 Deinking technologies

Plastic packaging is typically heavily printed with inks for functional benefits such as including information about composition, presence of allergens and nutritional details, etc., but also for marketing purposes to make them more appealing to consumers. The main constituents of printing inks are resins, solvents, colorants and additives. [32] Resins are high molecular weight polymers constituting 15 to 50% of the composition of the ink and they act as binder for colorant stabilization. [32, 33] Solvent constitutes the largest part of the ink composition (up to 65%). [32] Solvents are used to dissolve the resins and also to keep the ink liquid for supporting ink transfer. [34] Colorants used to give desired color to plastic packaging, constitute 5–30% of the ink composition. [32, 33] Colorants can be used as pigments or dyes. Pigments are insoluble solid fine particles which are dispersed in the binder, while dyes are substances that are completely soluble in the binder. [35, 36] In addition to colorants, lacquers or overprint varnishes are uncoloured forms of printing inks, which can be used to provide gloss and protective properties to the print. [33] Lastly, additives are generally used up to 10% in order to improve physicochemical properties of inks such as among, others, adhesion, slip and scratch resistance. [37, 38] The composition of these ink components can considerably differ depending on the printing process and also on the substrate. For example, for most substrates solvent-based inks in which the resin is dissolved in a suitable solvent are used as they allow sufficient wetting and adhesion. [34] Compared to solvents, the evaporation rate of water is much slower, making the drying process of the ink energy intensive. [34] Therefore, water-based inks are generally used for substrates which can promote absorption mechanism such as paper and board. [39] In addition to solvent and water-based inks where drying of inks is performed through evaporation of the liquid medium, in UV-based inks UV radiation is used for drying, which allows the
ink to immediately form a three-dimensionally cross-linked film. UV-based inks require reactive resins such as acrylates which can react with free radicals created by UV radiation. [33, 34]

Although inks are one of the necessary components of plastic packaging, they are a significant source of contamination in plastic recycling. As all printed plastic films are generally collected and processed together, a low quality brownish, grayish or black recyclate is obtained, making it only suitable for downcycled products. [40] The presence of ink also causes recycled films to be less stiff, weaker, and denser compared to the original material, thus its price is considerably lower than the price of films free of ink. Furthermore, during the processing or reprocessing, residual ink can also decompose and produce gases causing rancid odor formation and also decrease the physical properties of a raw material. [41] In order to eliminate these problems and obtain high quality recyclates, interest in deinking technologies is increasing. However, only a few deinking technologies are so far active in deinking of plastic packaging. For example, in the patented process known as the Nordenia Extraction and Cleaning process or NorEC (DE19651571A1), ethyl acetate was used as a solvent-based extraction medium to remove broad range of inks. [42] The NorEC process has currently been applied in an industrial packaging plant in the North of Germany with a capacity of 15000 tons. [42] In this plant industrial PE film waste is being shredded and treated with this extraction technology. It is stated that the NorEC process requires lower amount of energy compared to the conventional wet processing. [43] Furthermore, surfactants are extensively studied as a potential deinking medium. [44–48] Deinking mechanism by using a surfactant consists of four main steps: [1] adsorption of the surfactant on plastic surfaces, [2] solubilization of the binder in the surfactant aggregates so called micelles, [3] detachment of ink particles from the surface, and [4] stabilization of the detached ink particles (Figure 5). [48] According to the study of Chotipong et al. [48], cationic surfactants such as cetyl trimethylammonium bromide (CTAB) were more effective to remove both water- and solvent-based inks. In addition, it is shown that critical micelle concentrations (CMC), pH of the medium, temperature and stirring are important parameters on deinking efficiency. [48]

Furthermore, in the patented method of the University of Alicante (EP2832459B1), a surfactant is used to remove inks from plastic packaging. [49] This method has also a semi-industrial demonstration deinking plant with a treatment capacity of 100 kg/h. [49] In this closed-loop recycling plant, printed plastic films pass through several treatments such as grinding, deinking, washing, drying, and pelleting in order to obtain ink free plastics with high optical quality. [41] As a water-based medium is used, the medium can go to wastewater treatment. The use of surfactants to deink plastic films was also described in another patent filed by Duchenaud Uniflexo (EP1419829A1). [50] In this method, the deinking medium contains organic solvents and a non-ionic surfactant. Although high deinking efficiencies can be achieved via this method, it has been stated that physico-mechanical properties of recycling plastic substantially decrease. [41]

![Figure 5.](image)

*Four-step mechanism for removal of solvent-based ink from HDPE surface. Adopted from [48].*
dangerous products and the high cost of the process limit the potential scaling-up. [41] Similarly, the Italian company Gamma Meccanica uses mechanical brushes for deinking purposes, but again deinking is limited to packaging having the ink layer on the surface. [51]

2.4 Delamination technologies

Plastic packaging can be laminated with different polymer layers in order to increase their physico-chemical properties. Combination of functionality of each polymer provides plastic film with superior preservation performances such as among others, extended shelf life, high mechanical strength, good sealability (Figure 6). [52] For example, sealing properties of PET are poor, thus it is often laminated with polyolefins. [53] Similarly, use of aluminum layer provides protection against UV light, as such nutritional value of products are preserved for a longer time period. [53] Although the combination of different polymer layers extends the functionality and application area of plastic packaging, they make the recycling of multilayer packaging more complex. For example, during mechanical recycling, incompatibility issues may arise in the polymer blends due to their difference in physico-chemical properties, such as PE and PET. [54] Similarly, in thermochemical recycling heterogeneous polymers such as PET, polyamide (PA), polycarbonate (PC) contaminate polyolefinic plastic waste. [1] Therefore, multilayer plastic film fractions are still mainly incinerated or landfilled to date. [55] Recently, there is a growing interest towards single layer plastic films in order to eliminate those complexities encountered in multilayer packaging. [55] However, it is not always feasible to achieve functionalities that combination of different plastics can provide, thus in order to enhance circularity of flexible packaging, delamination of multilayer packaging might still be needed.

There are different option to separate the layers of multilayer plastic packaging. One of them is selective dissolution of a certain polymer layer. For example, in the study of Samori et al. [56] switchable hydrophilic solvents were used to selectively dissolve the LDPE layer and recover Al from food packaging. Similarly, in the study of Mumladze et al. [52] adhesive polymers were dissolved by using N, N-Dimethylcyclohexylamine (DMCHA) as a solvent and also during dissolution ultrasonic treatment was employed to speed up the separation of multilayers. Furthermore, in the patented method of Nauman and Lynch, pure polymer fractions were obtained through sequential dissolution of multilayer structures by gradually increasing temperature in the presence of a single solvent system. [57] There are several studies focusing on selective dissolution-precipitation of polymers to delaminate multilayer components, but they are mainly focused on recovering

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**Figure 6.**
An example of a multilayer flexible packaging film structure. Adopted from [6].
the polyolefins. [58–60] On the other hand, solvent-targeted recovery and precipitation (STRAP) process was studied to recover all the constituent polymer layers of multilayer plastic packaging. [61] This process would become competitive to design solvent systems for recycling of multilayer packaging. Another option to delaminate multilayer packaging is through selective decomposition of polymer layers. [62] For example, in the study of Kulkarni et al. [63], the aluminum layer was recovered from multilayer packaging structures by depolymerizing PET and PA in the presence of sub and supercritical water. In another study, sulfuric acid was used to degrade PET and recover the PE layer from multilayer structures consisting of PE and PET layers. [64] Although selective degradation of polymer layers are promising in terms of polyolefin recovery, degraded polymers affect the medium recovery adversely. It is shown in a study that during selective PET degradation, energy consumption for the solvent and product recovery contributes to a major part of the greenhouse gas emissions. [62] Multilayer packaging can also be separated through dissolution of tie layers such as among others, polyurethanes (PU), acrylates, acid anhydrides, or others which are used to laminate dissimilar polymer layers. There are several studies focusing on the dissolution of tie layers for separation of polymer-aluminum multilayer packaging by using organic solvent systems. [65, 66] For example, in the patented method of Panagiotis et al. [67] the cured composite laminate material was preconditioned to delaminate composite laminate materials by using organic solvents such as water, benzyl alcohol, acetone, methyl ethyl ketone (MEK), or a combination of one or more thereof. As an alternative to solvents, acids are also used dissolve tie layers towards delamination of a broader range of multilayer structures. [6, 68, 69] For example, in the patented method of Massura et al. [69] polymer, aluminum and/or paper were separated by using protonic carboxylic acids such as acetic acid together with organic solvents to increase the solubility of adhesives. In the study of Ügdüler et al. [6], it has been proven that diffusion rate of formic is faster compared to other longer chain carboxylic acids e.g. hexanoic acid, decanoic acid, thus formic acid was selected as a superior medium to delaminate different types multilayer packagings. Similarly, in various patents inorganic acids such as nitric acid and phosphoric acid are also used for delamination of composite packaging or industrial refuse containing aluminum layers. [70, 71]

To date industrial delamination technologies have mainly focused on tetra pack recycling. For instance, in China various companies are performing acid-based delamination. In this approach, recycling of composite packaging waste is carried out in a continuous industrial scale through separation of PE-Al by using formic acid and nitric acid. Afterwards, both materials are recovered by for instance a sink-float separation. [72, 73] In 2015, the German company Saperatec GmbH patented a method for recovery of saleable products from composite waste by using micro-emulsion comprising swelling agents, carboxylic acids, water, and surfactants. [74] Based on this patent, Saperatec has launched a project to build a recycling plant with a capacity of approximately 17000 t/a input. [75] The discussed delamination technologies are summarized in Table 2.

2.5 Solvent-based extraction methods

Solvent-based extraction methods can be applied to remove target additives from the polymer matrix without dissolving (e.g. Soxhlet extraction method, ultrasonic extraction method, etc.) or alternatively, (selective) dissolution can be applied to recover a certain polymer (from a mixture), which is typically called the dissolution-precipitation technique. In this chapter, solvent-based extraction methods will be divided into two groups: dissolution-precipitation and solid–liquid extraction methods.
2.5.1 Dissolution-precipitation technique

The dissolution-precipitation technique allows the removal of contaminants and additives from polymers. The principle of this technique is the dissolution of the polymer in a suitable solvent followed by solid–liquid separation steps for removal of contaminants. Next, the polymer can be recovered by rapid evaporation of the solvent or by adding a proper antisolvent that will make the polymer to precipitate. [77] Drying of the polymer grains and recovery of the solvent and antisolvents are the final steps. [78] Advantages of the dissolution of plastics in a suitable solvents are (i) the decrease in the bulk volume of the plastics, (ii) the precipitated polymer is in a more acceptable form for reuse, (iii) insoluble contaminants can be removed with a solid–liquid separation process and (iv) the final product might be competitive with virgin material in terms of quality. [79, 80]

Furthermore, dissolution-based processes can also be used for the separation of mixed plastic wastes based on the selective dissolution (Figure 7). [78] Selective dissolution-precipitation has been applied in a laboratory scale to different plastics, namely PP pipes, PVC bottles, PS waste foam, LDPE film and HDPE bottles from agrochemical packaging. [77] Pappa et al. [77] studied the selective dissolution-precipitation techniques at a laboratory scale and pilot-scale for the two-component mixture LDPE/PP. The solvent/antisolvent used was xylene/i-propanol in a 3:1 ratio and the dissolution was performed at different temperatures in the range of 85 to 135°C, depending on the polymer. The recovery of the two polymers was higher than 99%.

The choice of the solvent (S) and antisolvent (AS) is dependent on the solubility of the polymer and/or polymers. The amount of solvent used for dissolution also plays an important role, as low concentrated solution will lead to low viscous fluids, but will require higher amounts of antisolvent, since normally the added ratio of S/AS is 3:1. [81, 82] On the other hand, concentrated polymer solutions lead to very high viscous fluids, which are hard to process. [82] Therefore, typical recommended concentrations are in the range of 5–15 wt%. [81] Papaspyrides et al. [82] studied the dissolution-precipitation technique for LDPE pellets using xylene and toluene as solvents at 85°C. Toluene proved to be the most suitable solvent, as it permitted to
achieve higher concentrations (0.30 kg/L) and remaining within the limit of viscosity. The choice of the antisolvent is also important since it influences the form of the precipitated polymer. Some antisolvent may lead to gelly polymers while other permit the precipitation in the form of powder or grains. [78, 82] Papaspyrides et al. [82] concluded that acetone as antisolvent was the most successful one for the LDPE-toluene solution as the polymer was precipitated in the form of powder without forming gelling lumps. Table 3 summarizes solvent/antisolvent systems that have been proposed in literature for different polymer types.

Currently, there are some pilot plants on the market for plastics. The CreaSolv® technology, patented by Fraunhofer Institute IVV, consists of a dissolution-precipitation technique that is able to remove additives, for instance plasticizers, from different polymers, such as polyolefins, PS and PET in scrap packaging, among others. [87] The APK’s Newcycling® technology is designed to separate different polymer types such as PE and polyamide (PA) from multilayer plastic waste via a chemical dissolution process. [38, 88] The PureCycle TechnologiesSM process is designed to remove contaminants and purify PP, [89] and Polystyvert developed a dissolution process for recycling all types of PS. [90]

2.5.2 Solid – liquid extraction methods

Next to the conventional solid–liquid extraction (SLE) methods such as the shake-flask and Soxhlet extraction method, [91] alternative techniques like ultrasonic extraction, supercritical fluid extraction (SFE), microwave-assisted extraction (MAE) and accelerated solvent extraction (ASE®) have been developed. [91, 92] Compared to the traditional SLE, these techniques reduce the amount of solvent and shorten the residence time. [92] Further advantages of increasing the temperature and pressure during extraction is the performance enhancement due to the increase of solubility, mass transfer effects and disruption of the surface equilibria. [91] Table 4 summarizes the advantages and disadvantages of these techniques.
2.5.2.1 Conventional SLE: shake-flask extraction and Soxhlet

In a shake-flask extraction the polymer is mixed with the extraction solvent. The solvent will penetrate through the pores of the solid matrix and dissolve the polymer. [38] To speed up the extraction methods, the sample can be heated or refluxed. [91] After the extraction, the insoluble solid matrix can be separated from the solvent-containing analytes via decantation, filtration or centrifugation. [38, 91] The shake-flask method can be performed in batches and multiple extractions can be carried out. [91] The extraction efficiency is dependent on the type of solvent, extraction time, temperature, particle and pore size of the solid compound. [38] For example, Spell and Eddy [93] used the shake-flask technique to extract antioxidants (Ionol and Santonox) and a slip agent (oleamide) from PE at room temperature. For the extraction of these antioxidants with carbon disulphide, a time of 1–3 hours for Ionol and 50–72 h for Santonox at room temperature were necessary to achieve recoveries around 98%. Carbon tetrachloride was used for the extraction of the slip agent. After 21 h at room temperature, recoveries of 93 to 100% were obtained. [38, 94] The shake-flask method (which is basically a batch solid–liquid extraction) is a simple method, but it requires high residence

| Polymer | S/AS | Concentration | T (°C) | S/AS ratio | Polymer recovery (%) |
|---------|------|---------------|--------|------------|-----------------------|
| PET     | n-Methyl-2-pyrrolidone/n-octane [78] | 0.20 kg/L | 165     | 1:2        | 100                  |
|         | Benzyl alcohol/methanol [83] | 5%w/v | 180     | 1:3        | 99                   |
| PP      | Xylene/n-hexane [83] | 5%w/v | 140     | 1:3        | 98.7                 |
| PP      | Xylene/acetone [84] | 0.15 kg/L | 135     | 1:3        | 100                  |
| LDPE    | Xylene/n-hexane [83] | 5%w/v | 100     | 1:3        | 98.9                 |
|         | Toluene/Acetone [82] | 0.30 kg/L | 85      | 1:7        | ~100                |
| HDPE    | Toluene/acetone [85] | 0.10 kg/L | 110     | 1:4        | 100                  |
|         | Xylene/n-hexane [83] | 5%w/v | 100     | 1:3        | 98.6                 |
|         | Xylene/methanol [83] | 5%w/v | 100     | 1:3        | 97                   |
| PS      | Toluene/n-hexane [83] | 5%w/v | 25      | 1:3        | 87.7                 |
|         | Toluene/n-hexane [83] | 5%w/v | 50      | 1:3        | 92.1                 |
|         | Toluene/n-hexane [83] | 5%w/v | 100     | 1:3        | 94.5                 |
|         | Xylene/methanol [83] | 5%w/v | 25      | 1:3        | 89.2                 |
|         | Xylene/methanol [83] | 5%w/v | 50      | 1:3        | 95.8                 |
|         | Xylene/methanol [83] | 5%w/v | 100     | 1:3        | 97.9                 |
|         | Toluene/Water [80] | 0.50 kg/L | 75      | 1:3        | ~100                |
| PVC     | Cyclohexanone/n-hexane [86] | 0.30 kg/L | 25      | 1:7        | 95                   |
|         | Dichloromethane/methanol [83] | 5%w/v | 25      | 1:3        | 91.1                 |
|         | Dichloromethane/methanol [83] | 5%w/v | 40      | 1:3        | 98.2                 |
|         | Toluene/methanol [83] | 5%w/v | 25      | 1:3        | 94.1                 |
|         | Toluene/methanol [83] | 5%w/v | 50      | 1:3        | 94.6                 |

Table 3. Solvent/Antisolvent systems proposed in literature for several polymers typically found in plastic packaging waste streams.

Recent Advances in Pre-Treatment of Plastic Packaging Waste
DOI: http://dx.doi.org/10.5772/intechopen.99385
| Method                        | Principle                                  | Advantage                               | Disadvantage                                                                 | Industrial application                                                                 |
|-------------------------------|--------------------------------------------|-----------------------------------------|------------------------------------------------------------------------------|---------------------------------------------------------------------------------------|
| Shake-flask extraction        | Extraction with solvent                    | Low investment                          | Needs high residence time                                                    | Not yet proven for plastics, but is commonly applied in other sectors. [38, 95]      |
|                               |                                            | Simple equipment                        | Generally produces the lowest yields compared to the other extraction methods |                                                                                       |
| Soxhlet extraction            | Extraction with solvent at high T          | Low investment                          | Time and solvent consuming                                                    | Not yet available on the market for plastics, but commonly applied in other sectors, such as agriculture. [38, 100] |
|                               |                                            | Simple equipment                        | Relative selectivity                                                          |                                                                                       |
|                               |                                            | Automation                              | Requires more energy (compared to shake-flask)                               |                                                                                       |
|                               |                                            | High recovery                            | Potential for solvent loss                                                     |                                                                                       |
|                               |                                            | Excellent reproducibility                | Clean-up strategy required                                                    |                                                                                       |
|                               |                                            | No reverse diffusion of additives back into polymer | Less suitable for thermolabile analytes                                      |                                                                                       |
| Ultrasonic extraction         | Ultrasonic frequencies                     | Economical                              | Labour intensive                                                               | Still a challenge to scale up to industrial scale due to the high plastic volumes that might create engineering problems. [38] |
|                               |                                            | Simple equipment                        | Clean-up needed                                                               | Not yet on the market for plastics but applied in other sectors, such as extraction of caffeine. [38] |
|                               |                                            |                                         | No automation                                                                |                                                                                       |
| Microwave assisted extraction | Solvent with microwave absorbing component | Rapid                                    | Modest investment                                                             | Not yet available on the market for plastics, but commonly applied in other sectors, such as agriculture. [38, 104] |
|                               |                                            | Automation                              | Safety precautions                                                            |                                                                                       |
|                               |                                            | Low solvent use                          | Dielectric solvents                                                           |                                                                                       |
|                               |                                            | Simultaneous extraction                 | Requires optimization                                                         |                                                                                       |
| Supercritical fluids extraction | Use of supercritical fluids, e.g. scCO₂ | Reduced usage of organic solvent        | High investment                                                               | Not yet on the market for plastics, but applied to extract bioactive compounds in food industry. [38, 104] |
|                               |                                            | Shorter extraction time                  | Not eco-friendly                                                              |                                                                                       |
|                               |                                            | Adjustable solvent strength, Compared to Soxhlet, SCF can easily penetrate into a porous matrix | Requires high-purity extractants                                              |                                                                                       |
|                               |                                            |                                          | Requires optimization                                                         |                                                                                       |
...times. Currently, there are no such industrial plants within the plastic recycling sector, but it has been applied in other sectors. [38, 95]

The Soxhlet extraction method has been traditionally performed to extract polymer additives and residues with strong solvents at high temperatures. [96] The principle of the Soxhlet method is the continuous extraction of the solid target compound using a suitable solvent with repeated boiling-condensation cycles. [91] The extraction temperature is limited by the boiling point of the solvent used. [97] One of the disadvantages of this method is the time needed to extract the additives, that sometimes can go higher than 12 h. [98] For example, hindered amine light stabilizers (HALS) (Tinuvin 770 and Hostavin TMN 20) were extracted from a polyolefin with chloroform at 60°C for 16 h and around 96% recovery was obtained. [38, 99] The Soxhlet extraction method is not yet available on the market for plastics, but has been commonly applied in other sectors, such as agriculture. [38, 100]

2.5.2.2 Advanced solvent extraction methods: ultrasonic extraction, supercritical fluid extraction (SFE), microwave-assisted extraction (MAE) and accelerated solvent extraction (ASE®)

The principle of ultrasonic extraction is to detach the target compound by agitating and creating cavitation in the solid matrix using ultrasonic frequencies in the range of 20 to 2000 kHz. [38] Ultrasonic extraction is most typically performed using direct sonification by using an ultrasonic bath. [38] The parameters that are expected to influence the performance of ultrasonic extraction are temperature, particle size, solvent type and concentration, among others. [38] Haider and Karlsson [98] used the ultrasonic method to extract a HALS (Chimassorb 944) and two antioxidants (Irganox 1010 and Irgafos 168) from LDPE with chloroform at different temperatures (30 to 60°C) and times (5 to 60 min), obtaining recoveries of around 100%. [38, 98] This technique is not yet on the market for plastics but is applied in other sectors, such as extraction of caffeine. [38, 100, 101]

Supercritical fluids extraction enables the extraction of components from the polymer matrix by using a supercritical fluid, i.e. a substance above its critical temperature and pressure. [96, 102] A commonly used fluid is supercritical carbon dioxide (scCO2). [96, 102] The SFE process usually involves three steps: (i) diffusion of the solute from the core of the polymer to the surface, (ii) transfer of the compounds from the surface to the extraction fluid and (iii) elution of the compounds to the supercritical extractant. [97] The supercritical fluid extraction has several advantages over the Soxhlet extraction, these are (i) the use of less amount of solvent, (ii) shorter extraction time, (iii) adjustable solvent strength and (iv) a wider range of extraction temperatures, as it will not be limited by the boiling
Bermúdez et al. [96] used scCO$_2$ to extract 4,4 – dibutyl azobenzene from polystyrene matrices at 60–65°C and 22.4–24.5 MPa. The extraction yield improved by 41% using the SFE compared to the Soxhlet extraction [96]. Garde et al. [103] extracted phenolic antioxidants from PP using CO2 with hexane or methanol as a modifier at a temperature range of 30 to 60°C for 90 to 180 min and obtained recoveries higher than 80%. [38, 103] The supercritical fluid extraction is not yet on the market for plastics, but has been applied to extract bioactive compounds in food industry. [38, 104]

Microwave assisted extraction is based on the principle of using microwave energy in the range of 30 to GHz to 300 MHz to heat the solvent that is in contact with the solid sample. [91] The microwave energy is also used to partition the target compounds from the solid matrix into the solvent. [91] The use of polar solvents, such as water and acetone, is more advised for MAE compared to nonpolar solvents due to the low permittivity and thus, these would not be affected by the microwave energy. [38] Nielson [105] compared the extraction of antioxidants (BHT, Irganox 1010, Irganox 1076) and slip agents (erucamide) from PP, HDPE and LDPE with the Soxhlet and MAE technique. With the Soxhlet method, CYHA: 2-propanol and DCM: 2-propanol solvent mixtures at room temperature for 30 to 60 min were used and recoveries higher than 90% were obtained. [38, 105] With MAE at 48°C for 20 min Nielson [105] obtained recoveries higher than 90%. [38, 105] Costley et al. [106] used the Soxhlet method to extract cyclic trimer and other low molecular weight oligomers from PET with different solvents (xylene, DCM, acetone, water and hexane) at 140°C for 24 h. The recovery rates were around 60%. [38, 106] With MAE, Costley et al. [106] were also able to obtain recoveries around 60% at temperatures between 70 and 140°C for 30 to 120 min. Microwave assisted extraction is not yet available on the market for plastics, but has been commonly applied in other sectors, such as the agriculture sector. [38, 104]

Accelerated solvent extraction is a pressurized fluid extraction method. [38, 91] This technique is performed at elevated temperatures, usually between 50 and 200°C and pressure of 6.9 and 13.8 MPa, for a short period of time (5–10 min) and using low amounts of solvents (max 100 mL). [91] The principle of ASE® is the swelling of the polymer matrix by the solvent, followed by the desorption of the compounds from the solid matrix. Next, the diffusion through the solvent placed inside a particle core takes place and finally the transfer of the bulk of flowing fluid. [38] Garrido-López and Tena [107] extracted HALS and phenolic antioxidants from PE using 2-propanol with THF and/or CYHA swelling solvent at 80–110°C and 10.3 MPa for 2 to 22 min, obtaining recoveries higher than 97%. [38, 107] Vandenburg et al. [108] extracted a phenolic antioxidant from PP using 2-propanol at 150°C and 13.8 MPa for 5 min, obtaining a recovery of around 90%. [38, 108] The accelerated solvent extraction is not yet on the market for plastics, but has been applied in other sectors, e.g. in food and agriculture industries. [38, 104]

3. Conclusions

Plastic packaging waste streams are complex, typically containing several contaminants, additives and multilayer materials that complicate the recycling processes. Cleaning of plastic waste through advanced pre-treatment processes is a promising route to tackle the difficulties that are encountered in mechanical and chemical recycling due to contaminants.

Conventional pre-treatment steps that are currently applied in industries, which comprise sorting and water based washing, have shown to be not 100% efficient towards removal of the broad range of heterogeneous substances that are present on
a post-consumer plastic waste stream. Therefore, advances in pre-treatments have been proposed in literature, some already applied at pilot or industrial scale, whilst other are not yet on the market for the plastic recycling sector. Techniques such as deodorization and deinking have shown to be suitable for the removal of odors constituents and inks from plastics waste, respectively. Deodorization and deinking pilot plants are already available on the market but there are still some limitations to overcome. Delamination allows the separation of the different layers present in multilayer plastic packaging and currently, industrial delamination technologies have mainly focused on recycling of multilayers containing aluminum. Finally, solvent-based extraction techniques such as the dissolution-precipitation technique have shown to be able to remove target additives from the polymer matrix as well as to selectively recover different polymers. The dissolution-precipitation is already applied on the market for the removal of several contaminants, whilst other solvent based extraction techniques are not yet well established in the plastic recycling industry.

To conclude, there is still the need for further improvement and investigation to industrial implementation of several techniques for the plastic recycling sector. Nonetheless, advanced pre-treatment processes show a great potential towards upcycling and closed-loop recycling of plastics.

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Conflict of interest

The authors declare no conflict of interest.

Abbreviations

| Abbreviation | Definition |
|--------------|------------|
| AI           | Aluminum   |
| CAPEX        | Capital expenditures |
| CMC          | Critical micelle concentrations |
| CTAB         | Cetyl trimethylammonium bromide |
| DMCHA        | N, N-Dimethylcyclohexylamine |
| EPR          | Extended producer responsibility |
| HALS         | Hindered amine light stabilizer |
| HDPE         | High-density polyethylene |
| LDPE         | Low density polyethylene |
| MEK          | Methyl ethyl ketone |
| MRF          | Material recovery facility |
| NIR          | Near infrared |
| OPEX         | Operating expenditures |
| PA           | Polyamide |
| PC           | Polycarbonate |
PE Polyethylene.
PEG Poly(ethylene glycol).
PET Poly(ethylene terephthalate).
POPs Persistent organic pollutants.
PP Polypropylene.
PRO Producer responsibility organization.
PS Polystyrene.
PU Polyurethane.
PVC Poly(vinyl chloride).
VOC Volatile organic component.

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