## **Towards Higher Quality of Recycled Plastics**

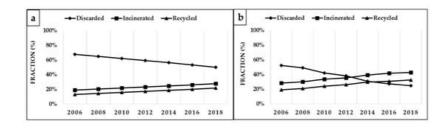
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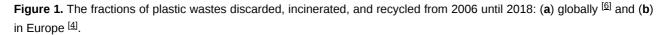
The increasing consumption of plastics and plastic products results in correspondingly substantial volumes of waste, which poses considerable environmental burdens. With the ongoing environmental actions, the application of circular economy on this waste stream is becoming inevitable. The quality of recycled plastic is generally determined by the homogeneity of the recovered plastic feed.

Keywords: plastic waste ; mechanical recycling ; recycled plastic quality

### 1. Introduction

Ending the plastic waste problem is becoming a global priority, pushing stakeholders to take actions to enhance the existing waste management systems and to invest in new ones <sup>[1]</sup>. Overall, recycling is the preferred option to treat plastics waste. However, when recycling is not the most sustainable option, energy recovery is the alternative <sup>[2]</sup>. All in all, both options complement each other to exploit the full potential of plastic waste <sup>[3]</sup>. The global amount of plastic waste incinerated and recycled is still low in comparison to plastic waste landfilling (see **Figure 1**a). On the other hand, Europe showed improvements in plastic waste. The same trend was experienced for mechanically recycled plastics from 2015 onwards <sup>[4]</sup> (**Figure 1**b). For the EU Countries with a landfill ban in place, thermally treated plastic waste still remains the most preferred management route <sup>[5]</sup>.





Plastic mechanical recycling can be performed in closed loops or open loops. Closed-loop recycling, also known as primary recycling, is the best to preserve the highest value of the material <sup>[I]</sup>. Only thermoplastics can be mechanically recycled with the possibility of closed-loop recycling <sup>[B]</sup>. Nevertheless, the plastic waste stream, particularly when it originates from private consumers <sup>[D][10]</sup>, is significantly heterogeneous and may contain hazardous substances hindering the application of closed-loop recycling <sup>[11][12]</sup>.

There are many additional factors affecting the plastic mechanical recycling practices, including separate collection schemes <sup>[13][14]</sup>, polymer type <sup>[15][16]</sup>, product design, and product category <sup>[17]</sup>. The different polymer types have different potentials for mechanical recycling. The chemical structure of the polymer and its chemical stability under recycling conditions define its recyclability and the number of recycling cycles it can endure. Additionally, different plastics have different applications and properties <sup>[18]</sup>. Hence, additives are differently utilised, as required by the material's quality or the product's design. The existence of chemical additives may interfere with the recycling process <sup>[11][19]</sup>. For instance, plastics containing volatile hazardous additives, such as brominated flame retardants, are not commonly considered for recycling <sup>[20]</sup>. The product design and product type may hinder recycling through the existence of multi-layer and multi-component materials in one product <sup>[9][21]</sup>. Rigid containers, consisting of a single polymer with minimum amounts of additives and inks, are more efficiently and economically recyclable in comparison to multi-layer and multi-component products <sup>[22]</sup>.

# 2. The Quality of Recycled Plastic Materials in the Context of a Circular Economy

Firstly, the definition of the term 'circular economy' is proposed by Kirchherr et al. [23]:

'A circular economy describes an economic system that is based on business models which replace the 'end-of-life concept with reducing, alternatively reusing, recycling and recovering materials in production/distribution and consumption processes, thus operating at the micro level (products, companies, consumers), meso level(eco-industrial parks) and macro level (city, region, nation and beyond), with the aim to accomplish sustainable development, which implies creating environmental quality, economic prosperity and social equity, to the benefit of current and future generations.'

An integral part of the circular economy for plastic waste is its recycling potential, which generally results from a trade-off between the quantity and the quality of the collected and further reprocessed material. In this sense, many factors are limiting the mechanical recycling of plastic waste  $\frac{[24][25][26][27]}{26}$ . Generally, to achieve the plastic waste recycling and recovery goals, sufficient quantities of plastic waste with adequate quality are required. Plastics quality deterioration can significantly limit the application of closed-loop recycling. Quality deterioration comes in the form of material degradation  $\frac{[21][28][29]}{28}$  and/or contamination  $\frac{[11][30][31][32]}{28}$ .

The degradation of plastic-based products typically takes place during plastic's (re)processing. Depending on the intended application, the degradation of the polymeric structure of a product occurs seldom as a consequence of its use. For instance, having a lifespan typically shorter than one year, post-consumer plastic packaging is less susceptible to degradation by use <sup>[33]</sup>. On the other hand, contamination may have different origins and is generally referring to polymeric sources—e.g., the case of polymeric cross-contamination—or foreign sources—as in the case of impurities or the presence of non-intentionally added substances (NIAS), resulting from the degradation of additives <sup>[34]</sup>.

Additional quality maintenance to the recycled materials can come in the form of precautionary actions, through identifying contaminants and their possible sources in the plastic waste fraction. This will provide a better-designed management system for plastic waste <sup>[34][35]</sup>. Consequently, the application of circular economy could be boosted in the plastic waste stream, while ensuring the sustainability, high quality, and safety of products <sup>[30]</sup>. Another factor to be considered is the heterogeneity of plastic products available on the market in response to their intended applications and design requirements <sup>[21]</sup>.

#### 2.1. Product's Designs

Plastics are made from an array of polymers, with combinations of different materials. Additionally, plastic's quality is modified by applying specific chemicals (e.g., colourants, additives, etc.) to meet manufacturers' functional and marketing demands <sup>[36]</sup>. Heterogenous designs obstruct the effective sorting of plastics, owing to the existence of countless colours, having different labelling materials that are hard to distinguish and remove, multi-layer and multi-component packages, etc. Additionally, the vast number of additives used in different products adds to the heterogeneity of the plastic waste stream <sup>[19]</sup>, resulting in an unfeasible and environmentally challenging recycling process, and consequently affecting the quality of recycled plastic <sup>[37]</sup>.

Currently, there is limited mechanical recycling of multi-layer and multi-component products, because of the crosscontamination between the different types and degrees of materials and polymers. In this case, the products' designs interfere with the closed-loop recycling of the material and prevent higher recycling quotas <sup>[22][38]</sup>. Next to the issues arising from the combination of different polymeric and non-polymeric materials, major limitations in the polymers' recyclability result to be inherent to the material, owing to its structural composition. In this sense, additives have proven to be a source of contamination generating organic substances, the behaviour of which is still under investigation <sup>[39][40]</sup>. The presence of these substances per se does not represent a recyclability burden. Nonetheless, the effects are exacerbated once they have to endure thermal processes, for instance, during extrusion or blow moulding.

#### 2.2. Collection and Sorting of the Waste Stream

Plastic waste collection and sorting are key steps in the mechanical recycling process <sup>[41]</sup>, as they come at the beginning of the mechanical recycling chain and consequently define the quantities and the complexity of the material sent for recycling. The current recycling schemes for post-consumer waste focus on an efficient and clean separation of post-consumer packaging waste (including PET water bottles and polyolefin packaging material) <sup>[42][43]</sup>. Collecting waste via

source separation waste collection systems is an essential part of increasing resource efficiency, achieving European recycling targets, and closing the loop in a circular economy <sup>[44]</sup>. The collection of plastic wastes can be conducted by bring-schemes, including the deposit refund system—DRS (e.g., for PET bottles) or through kerbside collection (plastic packaging material that is not included in the deposit systems) <sup>[22]</sup>. Many countries have a DRS in place targeting particularly PET bottles (in addition to glass bottles and aluminium cans), and they have proven high collection rates <sup>[45]</sup>.

#### 2.3. Contaminants

The primary sources of contaminants in plastics across the plastics' value chain were summarised by Pivnenko et al. <sup>[46]</sup>. They include additives, polymer cross-contamination, non-polymer impurities, and products generated by polymer degradation. Contamination occurs in all stages of the value chain except for the extraction process. Contamination starts with the production step (by additives). Most of the contamination occurs during the manufacturing step (due to design requirements: multi-layers, multi-components, labels, and inks) <sup>[47]</sup>. Further contamination occurs during the application stage (mostly non-polymer impurities as well as degradation products) <sup>[48]</sup>, segregation (polymer cross-contamination and non-polymer impurities), collection (polymer cross-contamination), sorting (polymer cross-contamination), and reprocessing (as a consequence of degradation) <sup>[49]</sup>. Among the numerous contaminants in plastics are the chemical additives that are added to provide predefined qualities for application and marketing needs <sup>[50][51][52]</sup>.

#### 2.4. Degradation

Changes in plastics properties (e.g., mechanical, optical, thermal characteristics) are a result of polymer degradation. As an example, cracking, erosion, discolouration, and phase separation might be the consequences of chemical transformation, bond scission, and/or the formation of new oxidised groups <sup>[28][53]</sup>. Degradation can occur via various pathways and by numerous factors. The reason behind plastic degradation can be attributed to: (I) the plastic composition, especially when the migration of additives produces irreversible tacking and warping phenomena; (II) ageing, which results in chemical instability over time; (III) environmental factors such as light, high energy radiation (UV, gamma radiation), microorganisms (i.e., bacteria or fungi), temperature, and humidity; and (IV) the improper usage and cleaning of the objects (see **Table 1**) <sup>[28][54][55][56][57]</sup>.

Type of Degradation or Decomposition	Degrading Agent
Photochemical degradation	Light (UV, visible light)
High energy radiation-induced degradation	X-rays, gamma rays, fast electrons
Photo-thermal or photochemical, ablative photo-degradation	Laser
Electrical ageing	Electrical field
Corrosive degradation, etching	Plasma
Biological degradation	Microorganisms
Mechanical degradation	Stress forces
Physical degradation, environmental stress, cracking	Abrasive forces
Ultrasonic degradation	Ultrasound
Chemical degradation or decomposition, etching, solvolysis, hydrolysis	Chemicals (acids, alkalis, salts, reactive gases, solvents, water)
Thermal degradation or decomposition	Heat
Oxidation, oxidative degradation and/or decomposition, ozonolysis	Oxygen, ozone
Thermo-oxidative degradation and/or decomposition, combustion	Heat and oxygen
Photo-oxidation	Light and oxygen

**Table 1.** Types of polymer degradation and the chief factors inducing degradation.

The types of degradation can be classified into abiotic and biotic degradation <sup>[58][59]</sup>. The rates of chemical and physical degradation of polymers are higher than those for biodegradation <sup>[60]</sup>.

Polymers can be subject to degradation mechanisms (otherwise erosion) within the bulk material or on its surface <sup>[61]</sup>. During the former, two main mechanisms occur: chain scission and thermodynamic changes in state. Specifically, chain scission occurs in the material, with a consequent decrease in the molecular weight and, therefore, in its mechanical strength. The mass of the polymer decreases infinitesimally and at a much slower rate until it disintegrates. Bulk degradation can occur due to thermodynamically unstable states in the material, such as internal stresses, imperfect crystallisation, limited compatibility of the used additives with the polymer or incomplete polymerisation processes. Contrarily, during surface erosion, the loss of material only takes place on the surface of the polymer, which undergoes a reduction in its size and mass. In this case, the molecular weight and mechanical strength of the polymer remain practically unchanged <sup>[61]</sup>. Typically, surface degradation occurs when the material comes in contact with the external environment.

The degradation of plastic can produce chemical changes as well. As a consequence of these changes, new functional groups in the polymeric chains are formed, resulting in the plastic's contamination and variation in the polymeric grades. These contaminants (oxygenated fragments) become trapped in the solid-state of the polymer and may diffuse through the melt, hindering an effective reprocessing and affecting the product's quality <sup>[54][38]</sup>. Under the environmental ageing condition, oxidation and hydrolysis are considered chemical degradation mechanisms changing the macromolecular properties of polymers <sup>[59]</sup>. According to the lifecycle periods of polymers, the relevant processes are classified as melt degradation, long-term heat ageing, and weathering <sup>[62]</sup>.

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