TECHNOLOGICAL CONSIDERATIONS REGARDING THE MECHANICAL RECYCLING OF WASTE FROM POLYETHYLENE AND POLYPROPYLENE PACKAGING

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Plastic materials have found applications in almost all fields as a result of their properties and low price. More than 30% of the production of plastics is intended for obtaining packaging. Since they are non-biodegradable and represent a hazard to the environment, strategies and directives have been adopted at national, European, and global level, regarding the recycling of packaging waste. The main ecological method of recycling them is mechanical recycling. The paper presents the main stages of an industrial mechanical recycling process, namely: collection, sorting, grinding, washing, drying, (purification), granulation, packaging, storage and marketing. This underlines the fact that in order to obtain high-performance PE or PP raw materials from recyclable polymer waste from packaging, the recycling process requires advanced sorting to separate them into polymer classes, because the mixtures of the polymers involved are incompatible in their melted state. This incompatibility leads to the lower processability and physical-mechanical performance of the products manufactured from these types of recycled polymer waste. In addition to the sorting methods currently used in the industry, new advanced methods of selective sorting of waste according to the type of polymer were also presented, such as: spectroscopic method, selective dissolution of polymers, thermal adhesion method, froth flotation method, electrostatic separation methods. It was emphasized that by using state-of-the-art technologies such as electron beam treatment followed by the electrostatic separation of waste mixtures from packaging, it is possible to obtain recycled polymers with high purity (90-97%) at advantageous production costs.

Keywords: plastic waste, PE, PP, packages, mechanical recycling, electrostatic separation

INTRODUCTION

Plastics have shown a significant development in the last 50 years, due to their high-performance properties that have allowed their use for a wide range of applications. In addition, they are light, cheap and can be easily processed into different products. The total amount of plastics generated in 2020 in the EU is 53.9 Mt, comprising: 45.7 million tonnes from plastics production (polymerisation) (about 84.8%), 4.6 million tonnes of post-consumer recycled plastics (about 8.5%) and 3.6 million tonnes of pre-consumer recycled plastics (about 6.7%). Plastics consumption in the EU in 2020 was 5.6 Mt and of this consumption, the largest amount (33.5%) is due to packaging. Because plastics, respectively plastic packaging are not biodegradable, and the end-of-life products from this class can pollute the environment, different strategies and directives have been adopted at national, European or worldwide level. Thus, the European Court of Auditors, through directive 94/62/EC on packaging and packaging waste, provides that the recycling rate of plastic packaging waste for 2025 to reach 50%, and for 2030 55%. The analysis of the 2006-2020 recycling evolution showed that the Compound Annual Growth Rate (CAGR) for this period was about 5.4%. Of the amount of plastics packaging waste collected in the EU in 2020 (17,946 kt), only 46% (8,171 kt) were recycled, 54% are still sent to energy recovery (6,689 kt) or landfill (3,085 kt). In Romania, in 2020, 44% were recycled from the collected plastic packaging waste (http://www.plasticseurope.org).

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The main types of polymers used to make the packaging are: low density (LDPE) polyethylene (PE) or high density (HDPE), polypropylene (PP), polyethylene terephthalate (PET), and other types. The main uses of these types of polymers (in virgin and recycled form, respectively) are presented in Table 1.

Table 1. The main types of polymers used to obtain packaging
(Kokkılıç *et al.*, 2022; Azeez, 2019; http://www.plasticseurope.org/)

Polymer	Percentage*	Applications of virgin plastic	Applications of recycled plastic
LDPE	17.4%	Reusable bags, trays and containers, agricultural film, food packaging film, wire and cable coatings, etc.	Trash bags, decking, shipping envelopes, housewares, buckets, wire and cable jacketing, carpet
HDPE	12.9%	Milk and detergent bottles, blowmolded beverage bottles, heavy-duty films, toys, utensils, housewares, pipe and processing equipment, wire and cable insulations, fibers for clothing, gas pipes, etc.	Decking, flower pots, crates, pipe, detergent bottles, food storage containers, swimming pool installation, corrosion protection for steel pipelines, folding chairs and tables, electrical and plumbing boxes, housewares, industrial wrapping and gas pipes etc.
PP	19.7%	Food packaging, sweet and snack wrappers, hinged caps, microwave containers, pipes, automotive parts, bottles, medical syringes, beakers, drinking straws, office furniture, clear bags, carpets, etc.	Plant pots, packaging articles, automotive parts, bottle tops, carpets, household components, 3D printing filament
PET	8.4%	Bottles for water, soft drinks, juices, cleaners, packaging film, soft drink bottles, other beverage, food & medicine containers, X-ray and photographic film, as fibers for clothing and carpets, staple fiber, etc.	Clothing, carpet, clamshell containers, water bottles, food packaging, house ware, lighting product, power tools, sports tools, water and soft drink bottles, thermally stabilized films, etc.

*Percentage of the total amount of plastics obtained in the EU, reported in 2020

Polyethylene (PE) is one of the most important polymers used in the production of packaging. In contrast to the large volume of packaging production, PE is one of the post-consumer recycled plastics capitalized in products with low use value. Consequently, there is a strong need to increase the recycling rate of the PE in order to manage and recover PE waste in an environmentally friendly way. The challenge lies in finding a cost-effective method for sorting, decontaminating food-grade PE, eliminating the hazardous impact caused by improper disposal, and stabilizing mechanical performance during the recycling process to ensure safe reuse and close the loop back into production.

The paper aims to present some aspects regarding the mechanical recycling of plastic waste, especially those from polyethylene and polypropylene packaging, and to bring up some considerations from a technical and technological point of view that could lead to the production of recycled plastics of high quality, with mechanical properties close to those of virgin polymers.

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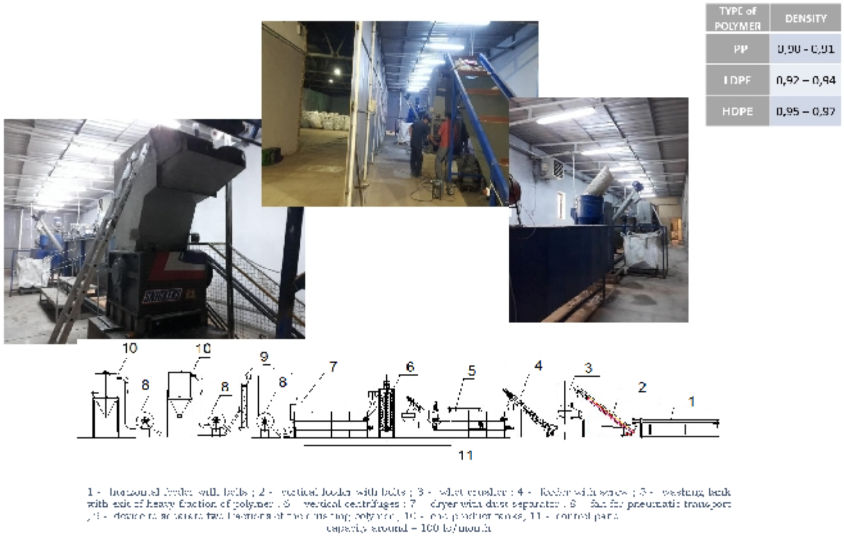
Industrial Technologies and Facilities for Mechanical Recycling of Plastic Waste

Mechanical recycling is the most important and environmentally friendly method of recovering recyclable plastics and consists of the processing of plastic waste into secondary raw material or products without significantly changing the chemical structure of the material. This recycling approach mainly includes a number of steps such as collection, sorting, washing and grinding of the plastic wastes (Al-Salem *et al*., 2009). Figure 1 shows the main structure of a "state-of-the-art" **mechanical recycling facility** for the recovery of recyclable plastics from sorted post-consumption packaging, from S.C. Monofil SRL, Săvinești, Romania. The first stage of the process is **the sorting** (manual or automatic) of recyclable plastics from non-recyclable plastics and other foreign substances. At the same time, the first grouping of waste according to the type of polymers is achieved. Manual sorting can be facilitated by material identification codes but the possibility of human error should not be neglected. Then follows the operation of **grinding** into flakes, followed by **washing and drying** the flakes (by dehydration and centrifuge). Dry flakes, if homogeneous, are directed to storage and then to processing by extrusion-granulation.



TYPE of POLYMER	DENSITY
PP	0,90 - 0,91
LDPE	0,92 – 0,94
HDPE	0,95 – 0,97

1 - horizontal feeder with belts ; 2 - vertical feeder with belts ; 3 - wet crusher ; 4 - feeder with screw ; 5 - washing tank with exit of heavy fraction of polymer ; 6 - vertical centrifuges ; 7 - dryer with dust separator ; 8 - fan for pneumatic transport ; 9 - [illegible] ; 10 - end product tanks, 11 - control panel
capacity around – 100 kg/hour

Figure 1. Technological flow and recycling facility of recyclable plastics from post-consumer packaging sorted from S.C. Monofil SRL

Certain types of materials such as HDPE or PET, require advanced purification in order to be used in the food industry, etc. This operation may include: chemical cleaning, optical sorting, washing for the label's removal, treatments at high temperatures and low pressures, etc.

The last stage is the **extrusion-granulation**, when the homogeneous waste flakes are melted and transformed into granules; in this operation can be added: plasticizers, antioxidants, compatibility agents, dyes, etc., to improve the properties of recycled waste. The recycled plastic material is obtained in the form of granules, which are then **packaged**, **stored** and **distributed** to consumers.

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Post-consumption packaging waste presents a series of **contaminations** that visibly affect the physico-mechanical characteristics and processability of the resulting recycled polymers. Of these, soluble mechanical impurities and dust are removed by classic processes in sorting and recycling facilities that operate on the principle of densimetric separation. Majority of the packages existing on the market are produced from two polymers, PE and PP, which have densities of less than 1g/cm^3, Table 2. The two polymers are immiscible in the molten state and therefore, the contamination of one of these polymers with the other (for example, HDPE contaminated with PP), leads to obtaining a recycled material with poor physico-mechanical and processing properties. The solutions adopted industrially for the removal of these contaminations consisted of:

1. Use of selective solvents for each of the mentioned polymers according to Table 2. This process is not cost-effective from an economic point of view due to the high costs as well as ecological in terms of solvent recovery.
2. Adding mineral additives in the technological process for the compatibility of the two immiscible polymers, PE and PP. This process was adopted by IMERYS for the production of HDPE for pipes that had a content of max 15% PP residual.
3. The use of polymers, such as LLDPE (linear LDPE), which ensures the compatibility of the two polymers at a concentration of maximum 10% PP in PE. This process is successfully used in the production of consumer goods and plastic pipes.

Table 2. Selected solvent in accordance with density

Polymer type	Polymer density	Solvent	Solvent density
Polypropylene (PP)	0.90-0.91	EtOH:H_2O (4:3)	0.92
Low-density polyethylene (LDPE)	0.92-0.94	EtOH:H_2O (1:1)	0.94
High-density polyethylene (HDPE)	0.95-0.97	H_2O	1.00
Polystyrene (PS)	1.05-1.07	10%NaCl/H_2O	1.08
Polyethylene terephthalate (PET)	1.38-1.39	$MgCl_2$ - saturated	1.34

Advanced Methods of Separation / Sorting of Waste According to the Type of Polymer

Certain types of polymers such as PP, LDPE and HDPE, are difficult to separate efficiently because of the small difference between their densities (Tall, 2000). In order to obtain high quality recycled plastic granules, numerous studies have been performed in which advanced methods of separation / sorting of waste have been successfully used, depending on the type of polymer. These methods are based on the properties of the basic polymers used in the packaging, which are presented in Table 3. A brief description of these advanced methods of sorting plastic waste is presented in Table 4.

Table 3. Physical proprieties according to each type of plastic
(Lim *et al*., 2022; Cho and Cho, 2020)

Plastic type	Thermal adhesion temperature, °C	Gravity specific	Work function, eV
LDPE	85-100	0.92-0.94	4.92
HDPE	120-140	0.95-0.97	4.77
PP	125-145	0.89-0.91	5.04
PS	105-140	1.05-1.07	4.22
PET	135-140	1.31-1.39	4.42

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Table 4. Advanced methods of polymer waste separation

Method	Description	References
Spectroscopic method	The spectroscopic methods include Fourier transform near-infrared spectroscopy (FT-NIR), mid-infrared spectroscopy (MIR), Raman spectroscopy, laser induced breakdown spectroscopy (LIBS) or X-ray fluorescence. They allow the separation of plastic materials with densities close in value.	Adarsh *et al*., 2022; Bonifazi *et al*., 2018
Selective dissolution of polymers	The solubility-based processes include stages of dissolving a series of incompatible polymers in a common solvent at various temperatures or in different solvents, so that one polymer is separated each time. These processes differ in the method employed to recover the polymer after the dissolution stage. The method requires high amount of energy and use of chemical solvents, is not considered sustainable.	Pappa *et al.*, 2001
Thermal adhesion method	The method using the different softening or melting temperatures of various types of plastics. LDPE can be separated from HDPE through this method because their thermal-adhesion temperatures are different (Table 3).	Tall, 2000; Lim *et al.*, 2022
Froth flotation method	Froth flotation is a physicochemical separation process based on the differences in surface properties of materials. The principle behind froth flotation is the attachment of the hydrophobic particles to air bubbles to be floated and recovered to the concentrate, while the hydrophilic particles are wetted by water and stay in the liquid phase	Kokkılıç *et al.*, 2022
Electrostatic separation	This technology makes it possible to separate different materials based on the differences between effective surface work function (Table 3).	Silveira *et al.*, 2018

Obtaining Recycled PE of High Purity by Electron Beam Treatment Followed by Electrostatic Separation of Packaging Waste Mixtures

Electrostatic separation methods allow obtaining materials of high purity (90-97%). This process offers high efficiency, low cost, and no concerns regarding secondary pollution (Wu *et al*., 2013; Silveira *et al*., 2018). It requires the waste to be dry and to have a size of 0.1-13 mm. In order to be able to apply this method, the waste flakes must be processed to acquire the electric charge. This stage can be achieved using: vibration, rotary drum, rotary blades, fluidized bed, cyclone, propeller-type tribo-charger, etc. Once the particles have acquired an electric charge, they can be separated electrostatically due to the difference between effective surface work function, Table 3. The most commonly used types of electrostatic separators are: the free-fall, roll, plate, and fluidized bed types. (Wu *et al.*, 2013). The separation process depends on many factors such as: residence time in the charger, air velocity, rotating speed, vibrating frequency, the relative humidity of the ambient in the charger, etc. For these reasons, it requires the optimal establishment of electrical and geometric parameters to allow efficient separation of waste.

In a study presented at the ICARS 2022 conference (Gosh *et al*., 2022) the charge induced by accelerated electrons was tested for the electrostatic separation of polyolefins, in a batch/discontinuous type procedure. It has been found that a pretreatment with low electron doses before electrostatic separation leads to specific charging of polymers, such as polyolefins, almost independently of additives and fillers (exempted talc). In the case of a 1-year with return on investment (ROI) and an annual flow rate > ~ 3750 t, the total price of sorted polymer materials amounts to less than ~ EUR 1000 / t and demonstrates that accelerated electron technology can provide cost-effective recycling systems for polymer waste. This procedure must be transferred to a continuous procedure so as to be integrated into a continuously operating thermoplastics recycling facility.

CONCLUSIONS

Plastics waste, and especially those from packaging, contain a mixture of polymers. For this reason, in the process of mechanical recycling of this waste, a sorting and separation by class of polymers is needed, in order to obtain recycled plastics of high quality, which could replace virgin plastics. Since the methods currently used in the industry need to be improved, several methods have been developed for the efficient separation of polymers from waste, but they are in an early stage. Of these new methods (spectroscopic method, selective dissolution of polymers, thermal adhesion method, froth flotation method, electrostatic separation methods), electrostatic separation methods allow obtaining materials of high purity (90-97%), and process offers high efficiency, low cost, and no concerns regarding secondary pollution. The method requires processing of the waste flakes to acquire the electric charge. Through a pre-treatment with low doses of electrons followed by electrostatic separation, an efficient separation of polymers from packaging waste (purity of over 90%) can be achieved in continuous flow and at low cost.

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