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Assessing the circularity of post-consumer HDPE milk bottles through open-loop recycling and their environmental impact

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ABSTRACT

Plastics are key in the packaging sector, but their widespread use contributes significantly to environmental challenges, such as the short life and high daily production of HDPE milk bottles. This study therefore aims to find a solution to this plastic waste, focusing on mechanical recycling. A comprehensive characterization of this post-consumer recycled HDPE reveals significant PP contamination, which poses a significant barrier due to polyolefin incompatibility, a common challenge in mixed plastics recycling. To mitigate this, blending with virgin HDPE and the use of various compatibilizers were investigated to improve the recyclability of the material. Several extrusion cycles were performed to analyse the thermo-mechanical degradation and to measure the performance and stability of the blends. The environmental impact of incorporating recycled HDPE into new bottles was also evaluated. Comparative evaluations with virgin bottles show that incorporating 25% or 50% recycled HDPE in the bottle yields carbon footprint reductions of 3% and 14%, respectively. These benefits could amplify with a wind-powered supply chain and a 100% recycled content. The findings lay the foundation for future plastic recycling scenarios, including dedicated sorting for this waste stream, providing a pathway to address the environmental impact of HDPE milk bottle disposal through recycling practices.

1. Introduction

In an ever-changing world, plastics stand out as materials with excellent physical and chemical properties, along with low production costs. In addition, the vast variety of polymers commercially available facilitate its use in different industrial sectors and products. These are all key elements that make plastics one of the World's most demanded material every year. According to the latest reports, 390 million tonnes of plastic resins were produced worldwide in 2021, being 57.2 of them synthesised in Europe (Plastics Europe, 2022). Whilst it is true that the production and demand have slightly decreased in the last year because of energy and logistics crises and the ongoing pandemic situation, the plastics industry remains as one of the most important, especially in the new challenges that societies are facing.

Among all the plastic resins that are currently used in the industry, polyolefins are the most abundant, thanks to their good mechanical properties and processability. Both polyethylene (PE) and polypropylene (PP) encompass around half of the European plastic demand in 2021 (Plastics Europe, 2022), available in a broad array of applications, including bags, toys, pipes, and specially packaging goods, being this sector the largest consumer of plastic resin with an estimated demand of 39.1% (Geyer, 2020; Westlie et al., 2022). However, this fact is also an important drawback for plastic materials, as most packaging products have single-use applications with short lifetimes, leading to large volumes of plastic waste produced annually.

Globally, plastic pollution has become a serious concern for ecosystems and habitats. The generation of plastic waste doubled between 2000 and 2019, reaching 353 million metric tonnes, according to a new report from the Organization for Economic Cooperation and Development (OECD) (OECD, 2022). Additionally, only 9% of these wastes are recycled, with most of them going to landfills (50%) or being incinerated (19%). To combat this issue, governments and organizations are

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promoting the recycling of plastic products through a battery of measures, including restrictions and taxes to reduce plastic' pollution (Clayton et al., 2021; Diana et al., 2022; Patrício Silva et al., 2020). However, OECD highlights that recycled polymers represent only 6% of total plastics production, and so there is much potential of improvement in this respect, especially for the purpose of achieving the goals outlined by the European Commission and implementing a Circular Economy in the plastics industry (European Commision, 2020; Commision, 2018).

Packaging products could be divided in those used in food contact applications (e.g., milk bottles) or non-food contact products (e.g., detergency bottles). While both types of containers are mainly made of high-density polyethylene (HDPE), their recycling rates currently differ. Most home and personal care plastic bottles are made up of a single layer of HDPE with similar properties despite their different manufacturers. Thus, these discarded goods are commonly mechanical recycled to obtain a resin that could be used again to manufacture other plastic products (Schyns and Shaver, 2021). Nevertheless, milk bottles must face some major challenges. First, milk containers could be designed in different ways, from one-layer packaging to a multi-layer bottle, usually with a barrier layer between the inner and outer (which contains titanium dioxide) that include additives like carbon black (CB) to maintain the organoleptic properties of sterilised milk (Johnson et al., 2015). Although results from other studies conclude that CB does not migrate from the matrix as itself (Bott et al., 2014), the possible presence of aromatic hydrocarbons and other substances could affect the recyclability of these plastic waste into food-contact applications. Moreover, the mechanical properties of these resins greatly vary from the grades used to manufacture detergency bottles, mainly their flexural modulus and environmental stress crack resistance, which makes it very valuable and necessary to separate both type of packages during the mechanical process to prevent contamination of the final recycled HDPE (rHDPE) grade (Kaiser et al., 2017; Ncube et al., 2021; Thoden van et al., 2021).

On the other hand, recycled polyolefins are not currently allowed for food contact applications by the existing standard due to lack of techniques to guarantee the effective decontamination of organic contaminants and other impurities (Cecon et al., 2021; Chen et al., 2021), except for closed-loop process where the origin of the material is well known, in particular the scrap and cut-off from post-industrial lines (Su et al., 2021; Welle, 2005). This issue, together with the presence of CB, ends up limiting the reutilization of rHDPE recovered from milk bottles, especially if the target is to reuse them in the same application, i.e., closed-loop recycling. Another obstacle for this recyclate is the contamination with other polymers during the recycling process, including the presence of other HDPE grades from different applications. Current sorting methods in the recycling plants include direct sorting such as magnetic or floating separation (Serranti et al., 2015), or indirect sorting based mainly on spectroscopic techniques, mainly infrared spectroscopy. While various mathematical approaches report an accuracy of 95% when employing near-infrared spectroscopy (NIR) (Madan Kumar et al., 2014), this methodology may be affected by the surface roughness, or moisture in the sample, affecting the separation efficiency (Duan and Li, 2021; Küppers et al., 2019) Thus, in this particular case, the presence of other polymers is a common phenomenon, as the bottle caps are mainly made of PP, which implies the presence of moderate but harmful PP contents in the rHDPE resins. This is an important consideration since the incompatibility among both resins leads to a material with poor mechanical properties, thus limiting the obtention of high-quality recyclates and being the main factor that could restrict their incorporation in the industry (Karaagac et al., 2021a; Gall et al., 2020).

To improve the performance of these recycled resins, several solutions have been explored, predominantly the use of compatibilizers that could help to enhance the adhesion between both polyolefins (Blom et al., 1998; Klimovica et al., 2020; Sjoqvist and Boldizar, 2011; Xu et al., 2018). However, when investigating the usage of compatibilizing agents, it is important to consider how their addition affects the mechanical properties of the materials and, consequently, increases costs. Other possibility that can be studied is blending the rHDPE with virgin resins to avoid the loss of properties when using a recycled plastic (Juan et al., 2020, 2021a).

This work therefore focuses on the evaluation of rHDPE made from post-consumer discarded milk bottles while exploring potential approaches to improve the qualities of this material, trying to increase the recycling rates of this flow and reach the target of recycling 55% of plastic packaging waste by 2030 (Commision, 2018). It is important to note that the aim of this study is not food safety, but rather to provide valuable insights into how to improve the circularity of polyolefin plastics by addressing the physico-chemical challenges associated with this particular recycled plastic flow, and thus gain knowledge for potential applications in the plastic industry, including dedicated sorting for this stream. As a research object, this study focuses specifically on Spanish HDPE milk bottles, collected, and sorted to avoid contamination with other plastic bottles or packaging materials, such as beverage cartons or PET bottles. To evaluate the effects on mechanical performance, various commercially available compatibilizers as well as blends with virgin resins have been employed. The main properties evaluated, considering a bottle to bottle open-loop scenario, are short-term properties like flexural modulus, toughness, impact strength, micromorphological characteristics and environmental stress cracking resistance (ESCR), a key property in HDPE packaging that is more sensitive to the recycled content due to the presence of impurities (Freudenthaler et al., 2023). In this scenario, replacing part of the virgin resin with recycled resin has clear environmental benefits. This substitution plays a key role in reducing the environmental impacts linked to the manufacture of plastics, while facilitating the integration of recycled resin into the market. Blends containing 50% by weight of recycled and virgin milk bottle material, were subjected to up to 10 reprocessing cycles to provide a comprehensive degradation analysis. A life cycle assessment (LCA) was then conducted to assess the potential environmental benefits of including recycled material in new HDPE bottles. Different scenarios were investigated, testing the environmental influence of the recycled material incorporated in the bottle and the electricity source used throughout the supply chain.

2. Material and methods

2.1. Materials and blends preparation

The rHDPE for the purposes of this study was obtained from post-consumer milk bottles provided by a Spanish recycler, referred to as rPE in this research. The HDPE milk bottles commonly found in Spain are mainly multi-layer HDPE bottles (Fig. S1 in Supplementary Information), with an intermediate layer containing carbon black and a content lower than 1% (EN ISO 53375-2). The label and cap are mainly made of PP, so it is important to separate both parts from the bottle to prevent contamination of the recovered HDPE.

The post-consumer milk bottles were collected from the Spanish lightweight packaging waste fraction. These lightweight packaging were transported to a material recovery facility where they were separated from other end-of-life products and sorted by type. Then, the HDPE were transported again to a recycler industrial plant, where the milk bottles were separated from other HDPE bottles, such as detergency bottles, as they are usually made from different HDPE grades. Next, the material underwent a mechanical recycling process consisting of shredding, floating separation and cold-water washing steps to reduce the presence of undesirable substances (labels and organic contaminants), a drying process by centrifugation with air, and finally a melting and extrusion process to homogenize the material and obtain the recycled pellets.

Once the recycled material was obtained and characterized, it was blended in a Collin ZK25 twin-screw extruder with virgin resin at various recycled levels (up to 75% of rPE). The extrusion temperature ranges from 160 to 220 °C, while the screw speed was set to 60 rpm. The virgin resin chosen for blending with the recycled material is a blow-moulding commercial grade supplied by Repsol Spanish company, like the ones used to produce HDPE milk bottles.

Different commercial compatibilizers were chosen to evaluate their effect on the rHDPE. Five different additives were tested, two elastomeric polyethylene (PE) (Fusabond N215, named C1 and Versify 2200, named C3), maleic anhydride grafted PE (Fusabond E100LG, named C2 and Fusabond E204, named C5) and ethylene-copolymers (Entira EP1753, named C4), all of them supplied by Dow Chemical Company.

2.2. Molecular and physical characterization

Density and melt flow index (MFI), both fundamental thermophysical properties of polymer materials, have been determined for all HDPE materials and its blends. To evaluate the density the immersion method was followed, according to the procedure described in the UNE-EN ISO 1183-1 standard. Melt flow rate was measured using a CEAST tester in which, following the UNE-EN ISO 1133-1 standard, between 6 and 7 g of sample are heated at 190 °C. Once the sample is melted, a weight is applied (2.16 kg) and an extrudate is obtained, which is weighed to calculate the amount of material that flow in 10 min. Thermal measurements were performed in a DSC Mettler-Toledo 822e with a heat rate of 10 °C/min.

A CRYSTAF-TREF equipment model 300 from Polymer Char was used to evaluate the PP content in the recycled material. Fractionation techniques, such as Temperature rising elution fractionation (TREF), proved to be an effective technique for the identification and quantification of PP impurities in this kind of rHDPE resin (Juan et al., 2021b). Around 80 mg of sample was dissolved in 1,2,4-trichlorobenzene at high temperature (160 °C), after which it was loaded into the column and cooled to 35 °C at a constant cooling rate of 0.5 °C/min. After the crystallization step, the sample was eluted at a constant rate of 1 °C/min, measuring the concentration of the polymer with an infrared detector.

2.3. Mechanical characterization

Mechanical properties were assessed for all samples and blends at 23 °C and 50% relative humidity. In accordance with to UNE-EN ISO 527-2, the dumbbell-shaped specimens 1BA were subjected to tensile tests using a universal testing machine (MTS Alliance RT/5). Flexural modulus and Charpy impact resistance were measured on prisms (80 mm × 10 mm × 4 mm), following the ISO 178 and UNE-EN ISO 179-1, respectively.

Hardness has also been determined since it is a property that indicates the resistance that the material opposes to being scratched or penetrated. The Shore D hardness has been determined according to the UNE-EN ISO 868 standard. An AMSLER durometer was used on a 4 mm thick test specimen for 15 s of application.

The resistance to environmental stress cracking (ESCR), a critical property for HDPE bottles subjected to stress under a chemical agent or environmental stress, is evaluated through the Bell Telephone Test (condition B of ASTM D 1693) at a temperature of 50 °C.

Finally, SEM pictures were captured from the fracture surface of Charpy impact strength samples, using a Philips XL-30 ESEM.

2.4. Life cycle assessment

The previously generated knowledge was compiled to assess the potential environmental benefits of incorporating recycled material into new non-food HDPE bottles. An attributional LCA (International Organization for Standardization, 2006a; International Organization for Standardization, 2006b) has been performed to evaluate two main scenarios, which involved incorporating 25% or 50% of recycled material into the bottle. A hypothetical scenario in which the bottle is manufactured with 100% recycled material was also analysed assuming advancements in recycling technologies and regulatory conditions. This scenario can be optimal for certain applications that tolerate or even benefit from higher recycled content, e.g. for packaging low-concentrated detergents (OECD, 2021). In order to further assess the influence of electricity impacts throughout the bottle's supply chain, the three scenarios were evaluated considering either the average Spanish grid mix or on-shore wind power (as an example of a low-carbon power generation option). All scenarios were compared against the production of a 100% virgin HDPE bottle (i.e., 0% recycled content) to assess their potential environmental benefits. Given that our LCA aims to provide insights into the environmental performance of alternative scenarios for producing an HDPE bottle, our study aligns with Situation A: "Micro-level decision support", as defined in the International Reference Life Cycle Data System (ILCD). According to the ILCD, the attributional approach using average market data is the most suitable for life cycle inventory (LCI) modelling in this context.

The functional unit analysed is the production of a standard single-use non-food plastic bottle that has a volume of 1.5 L and an average weight of 49.4 g. The system boundaries are cradle-to-gate, covering all the relevant impacts from virgin materials acquisition to the production of the bottle. The use and end-of-life stages have not been considered as these stages are assumed to be the same in the scenarios, meaning that they do not affect the comparative analysis. The processes considered for the supply of rHDPE are the collection and transportation of post-consumer HDPE milk bottles, mechanical sorting at a material recovery facility (MRF), transportation to the recycling facility, treatment of rejects, and recycling into HDPE flakes (Fig. 1). It should be noted that the post-consumer HDPE milk bottles are considered burdens free, i.e., they do not bear any impacts from the previous life cycle. This so-called cut-off approach was applied to be consistent with the assumption in the ecoinvent database, which was used as background LCI data source (see below). The processes considered for the supply of fossil HDPE cover from the supply and steam cracking of naphtha to the production of virgin HDPE granulate. Moreover, the processes for the production of non-food bottle-grade HDPE granulate through extrusion, compatibilizer additive, and the bottle via blow moulding are considered within the system boundaries.

The LCI for all the proposed scenarios, available in Table S1 in the Supplementary Information, was prepared based on the experimental work and previous research from the authors (Istrate et al., 2021). The post-consumer HDPE milk bottle is collected by the MSW collection service and transported by a diesel truck over 30 km to a MRF (Istrate et al., 2021), where HDPE is mechanically sorted with an efficiency of 85% (Antonopoulos et al., 2021). The MRF consumes electricity and diesel for machineries operation. The recovered material is then baled and transported over 300 km to the recycling facility. Here, the HDPE is mechanically recycled into flakes with an overall efficiency of 88% (Antonopoulos et al., 2021). This process requires electricity, diesel, heat, and water and chemicals for washing. The recycled HDPE flakes are blended in an extruder with virgin HDPE to produce non-food bottle-grade HDPE granulate with an efficiency of 98% (Garcia-Gutierrez et al., 2023). The recycled content of the obtained granulate is specific to each scenario (i.e., 25%, 50%, or 100% recycled content). As for modelling the LCI for virgin HDPE, the global market dataset available in the ecoinvent v3.8 database was used without further modifications. This dataset represents the global average production of virgin HDPE, encompassing the following region contributions: 56% Asia and the Pacific, 19% Europe, 18% North America, 4% Africa and 3% South America. The extrusion process consumes mainly electricity and heat. Moreover, 3% of an elastomeric PE compatibilizer (made from 75% PE and 25% synthetic rubber) is added. Finally, the HDPE bottle is produced via blow moulding, while the production of the bottle seal (0.4 g of aluminium), cap (4.3 g of PP), and label (0.8 g of low-density polyethylene) is also considered. Inventory data for background processes (e.g., electricity mixes) was obtained from the ecoinvent v3.8 database (cut-off system model). The Brightway2 modelling software (Mutel, 2017) was used to implement the LCIs and to conduct the LCA.

The 16 impact categories included in the Environmental Footprint

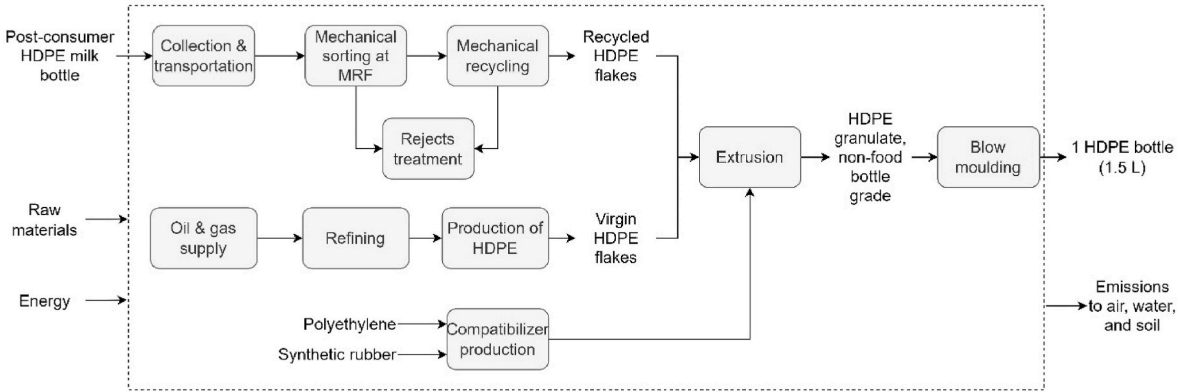

Fig. 1. System boundaries for the production of a standard single-use HDPE bottle from recycled and virgin HDPE.

(EF) method recommended by the European Commission were assessed (Fazio et al., 2018). These categories include climate change, acidification, eutrophication (freshwater, marine, and terrestrial), photochemical oxidant formation, particulate matter formation, ozone depletion, ecotoxicity, human toxicity (carcinogenic and non-carcinogenic), ionizing radiation, and resources use (non-renewable, metals and minerals, land, and water). Climate change impacts were assessed with the IPCC 2021 method considering a 100-year time horizon (Masson-Delmotte et al., 2021). The other impact categories were assessed with the methods recommended in the EF v3.1 (Fazio et al., 2018).

3. Results and discussion

3.1. Analysis of post-consumer milk bottles, rPE

In this study, the properties of post-consumer rHDPE recovered from milk bottles (rPE) have been evaluated. To enhance the properties of the rHDPE, the approach used herein consisted firstly in mixing rPE with a commercial HDPE virgin resin proceeding from blow moulding, and eventually with compatibilizing agents to solve the contamination concerns. Table 1 summarizes the main properties of the polymeric resins that have been used throughout this research work.

First, rPE was deeply analysed by different techniques. As for of density, compared to the virgin resin, there is a clear increment that could be easily explained as the recycled material contain fillers and pigments added during the manufacturing (substances such as carbon black to protect against UV rays or calcium stearate as a lubricant and release agent) (Wiesinger et al., 2021). Additionally, there were significant differences in the MI between virgin HDPE and rPE, which is a likely indicator that the recycled material is contaminated with other polymers. As contamination is a common phenomenon during mechanical recycling of polymers, especially in polyolefins, DSC and TREF analysis were performed to determine the occurrence of undesired compounds in the rHDPE.

Melting thermograms of virgin and recycled resins by DSC are shown in Fig. 2 (a). Between 135 and 140 °C a sharp peak is observed, which corresponds to HDPE. However, for the rPE, a shoulder around 160 °C is also noticed. This signal is indicative of the possible presence of a certain amount of PP in this sample. However, as demonstrated by other research (Juan et al., 2021b), this approach is inaccurate in quantifying lower levels of PP impurities in these polyolefin blends, resulting in an underestimation of the quantities of PP in these recyclates (Pasch et al., 2000). Therefore, an alternative technique based on TREF analysis were also performed to determine the actual content of these impurities more precisely. Fig. 2 (b) shows that TREF thermograms for polyolefin blends show two distinct peaks, one at 95–100 °C and the other at 115–120 °C, which correspond to the PE and PP, respectively (Fernández et al., 2015). For the virgin PE, only one peak is observed around 100 °C, which is assigned to HDPE. Nevertheless, two peaks could be observed for rPE, at 98 °C and 120 °C, which undoubtedly confirms the presence of PP. Moreover, from this result the PP content could be estimated to be around 5%. These PP contamination levels measured for this recycled resin are far from the 1–2% of PP that is usually admitted and reported for recycled PP's, which makes it necessary to look for solutions to improve the properties of this recyclate and to shed more light on this post-consumer plastic waste that is managed by municipalities across Europe.

3.2. Effect of compatibilizing agents

The immiscibility of PE and PP causes interfacial tension resulting in poor mechanical properties and a reduction in both tensile strength and impact strength features (Kukaleva et al., 2003; Van Belle et al., 2020; Greco et al., 1980). For the compatibilization of polymer mixtures, compatibilizing agents have been explored in the past as a possible solution to improve the compatibility between HDPE and PP. Those agents consist mainly in block or graft copolymers (Vervoort et al., 2018; Teh et al., 1994; Karaagac et al., 2021b).

Thus, as a possible solution to improve the mechanical properties of rHDPE, five different compatibilizing agents have been tested (C1 to C5). Each one of these agents have been added at 6 wt% to a control blend (called M), previously prepared, which contains 95% of virgin HDPE and 5% of PP. The effect of these additives on key properties in plastic bottle design like flexural modulus and impact resistance were studied. Results are summarized in Fig. 3.

Table 1
Physicochemical properties of virgin and recycled HDPE.

Material	Type	Density (kg/m^3)	Melt Index (g/10 min)	Flexural Modulus (MPa)	Impact Resistance (kJ/m^2)	Hardness (Shore D)	ESCR (h)
PE	Blow-moulding	962	0.26	1540	25	65	15
rPE	Post-consumer Milk Bottles	982	1.00	1311	3.1	61	12
pi-PE	Post-industrial Milk Bottles	981	0.31	1498	15	64	12
pi-PP	Post-industrial Milk Bottles Caps	983	59.5	1885	1.8	71	–

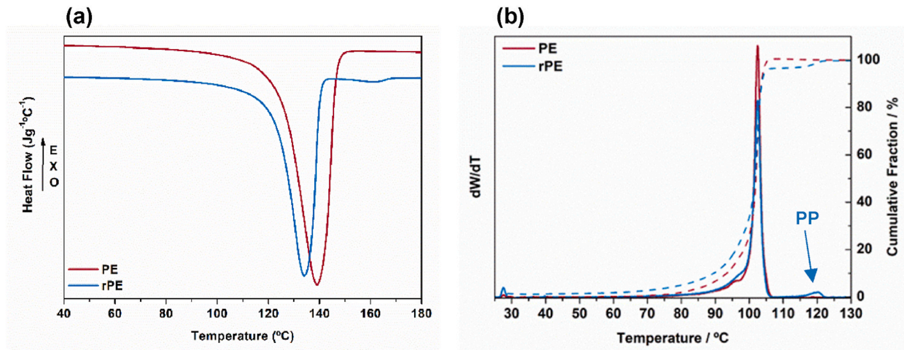

Fig. 2. DSC melting curves from second heating scanning of virgin and recycled resin (a) and Analytical TREF curves for both virgin and rHDPE (b).

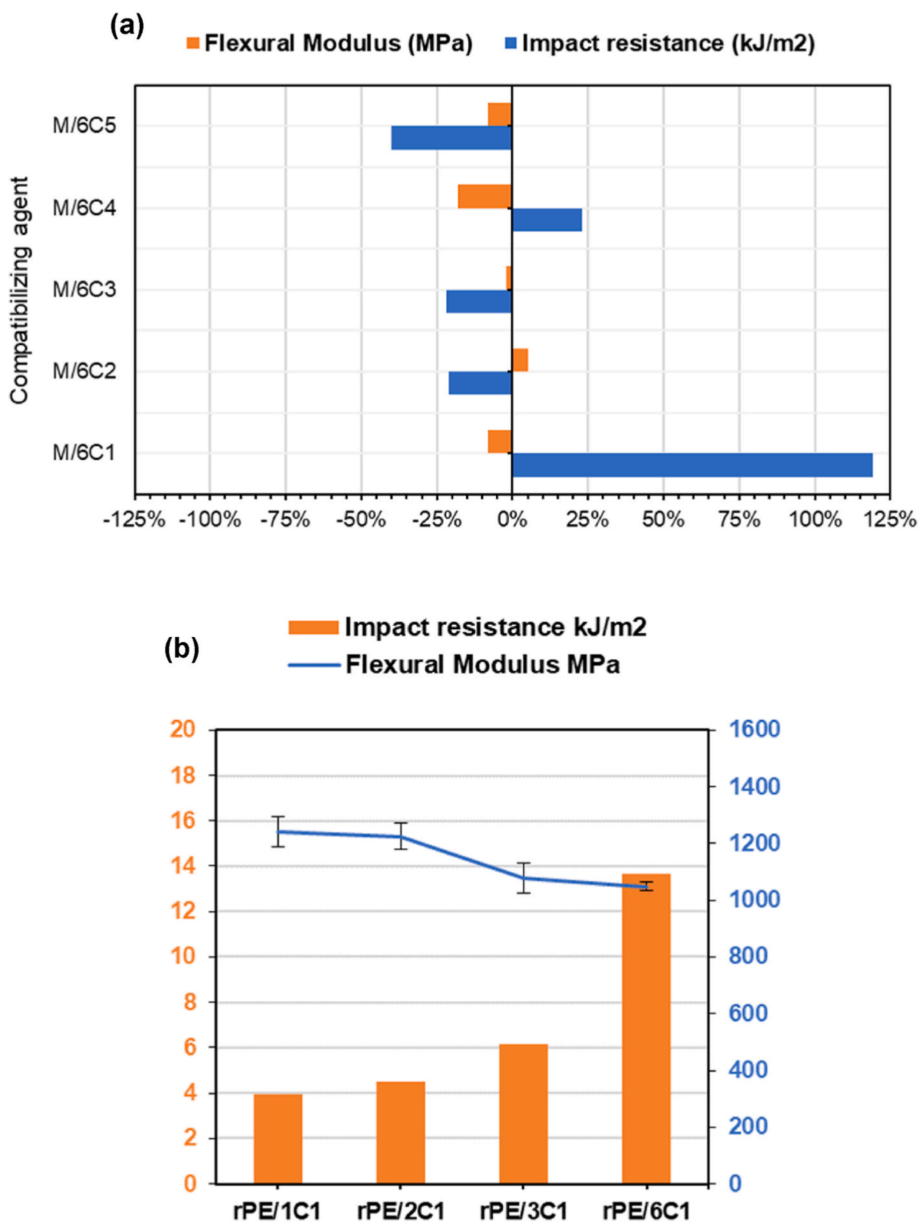

Fig. 3. Variation of mechanical properties for reference blend after adding 6 wt% of each compatibilizing agent studied (a) and mechanical properties of rPE adding compatibilizer C1 at different contents (b).

When comparing the five compatibilizing agents results, it can be seen an improvement in the impact strength resistance only for the blends containing C1 and C4, while for the rest of the agents tested this property decreased. However, the flexural modulus, another critical property in plastic bottles, was more sensitive to the presence of C4, while when C1 is chosen, the flexural modulus is less affected as shown in Fig. 3 (a). Therefore, these results conclude that the most suitable compatibilizing agent for this study is C1, although the ratio should be adjusted to optimize the properties of the blend.

Once the C1 compatibilizing agent have been chosen, it was tested on the recycled post-consumer bottle at different percentages to establish the optimal content of C1. Fig. 3 (b) displays the four blend systems that were prepared, containing 1, 2, 3 and 6 wt% of compatibilizer. Results obtained show that the impact strength resistance increases significantly when 6 wt% of C1 is added. This was envisaged due to the elastomeric features of the compatibilizer. Similarly, the flexural modulus decreases with the increasing content of the compabilizing agent. This is especially noticeable when 3 wt% of C1 is used, as a reduction of approximately 200 MPa is observed. To prevent a higher drop of the flexural modulus, 3 wt% of compatibilizing agent has been determined to be the appropriate content for this specific case of study, as the addition of 6% by weight of compatibilizer could affect the stiffness of the bottle and considerably raise the cost of the process. Furthermore, 1 and 2 wt% were discarded, as the impact resistance values barely increase.

3.3. Effect of PP contamination in recycled HDPE

Following the initial characterization of the rHDPE, the existence of slight but meaningful amounts of PP in these recycled resins could be associated to the presence of caps or pieces of them in the recycled streams. Both polyolefins are difficult to separate due to similarities in densities, which makes that small amounts of PP are commonly found in rHDPE resins. In this sense, the plastic waste pollution and the related environmental problems has led to the adoption of new measures and policies by the EU. In 2024, as part of the Single-Use Plastic Directive of the EU (European Parliament and the Council of the European Union, 2019) it will be mandatory for caps and lids to remain attached to all drinking packaging with a capacity up to 3 L, hindering the separation of the cap from the bottle.

For these reasons, considering that under this scenario rHDPE is susceptible to contain higher amounts of PP from caps, preparation of three blends at different PP contamination levels has been carried out. To prepare these blends and simulate the milk bottle and the cap, scrap obtained from milk bottles and caps manufacture (pi-PE and pi-PP, respectively) were used, the properties of which are described in Table 1.

Since the presence of caps and other materials could vary, as well as the efficiency of the sorting and separation process, blends with 2.5, 5, and 7.5 wt% of PP were prepared, simulating different theoretical scenarios of contamination. The 5 wt% contamination was evaluated due to the presence of PP previously determined in the recycled post-consumer milk bottle. On the other hand, assuming that the cap is bound together with the plastic bottle, an average content of 7.5 wt% of PP is expected, as the PP represents around 6–9 wt% of the common multilayer milk bottles found in the Spanish market. Finally, 2.5 wt% is a hypothetical case where an optimal separation process during mechanical recycling would have taken place.

To assess the performance of HDPE in the presence of various PP amounts, the mechanical properties of these blends were thoroughly studied (Fig. 4 (a)). Results shows that an increase of PP in the PE matrix caused a drop in the Charpy impact strength resistance especially for the sample contaminated with 7.5 wt% of PP, while flexural modulus remains unchanged. The key factor explaining this behavior is the immiscibility of the two polymers since the poor adherence of the interfaces embrittles the blend and results in subpar mechanical performance. This result proves that it is essential a better eco-design of the plastic packaging and better sorting and classification technologies to prevent that contamination with PP caps increase the presence of this polyolefin in rHDPE sources, leading to a poorer quality recyclates.

The compatibilizing agent C1 were also tested in these blends, at a concentration of 3 wt%. As seen in Fig. 4 (b), the impact strength resistance is greatly affected by the presence of the compatibilizer, doubling the values obtained for each sample. However, the drawback of the addition of this additive to the blend is the decrease of the flexural modulus, as it exhibits a decline between 100 and 200 MPa, which means that the bottle is less rigid, although still in the acceptable range of flexural modulus for milk bottle application. Therefore, the plasticizing effect of compatibilizers must be consider when designing the product, even though their use may be a way to mitigate to some extent the negative effects caused by the existence of PP in the recycled material.

3.4. Post-consumer HDPE milk bottles and circular economy

3.4.1. Assessment of the circularity of post-consumer milk bottles

Considering all the results obtained during the characterization of the rHDPE from milk bottles, the final part of this study assesses the possibility of reintroducing this recycled flow again in the plastics industry in a bottle-to-bottle open-loop recycling scenario. Two distinct strategies were put forth to enhance the recycled resin's characteristics. On the one hand, rPE was mixed with virgin resin at different contents (25, 50 and 75 wt% of PE) to evaluate the effect on its mechanical properties. On the other hand, to reduce the negative effects of the PP, compatibilizing agent (C1) was added during the blending process. All blends prepared for both scenarios were extensively characterized, focusing on the physical properties, as well as the mechanical

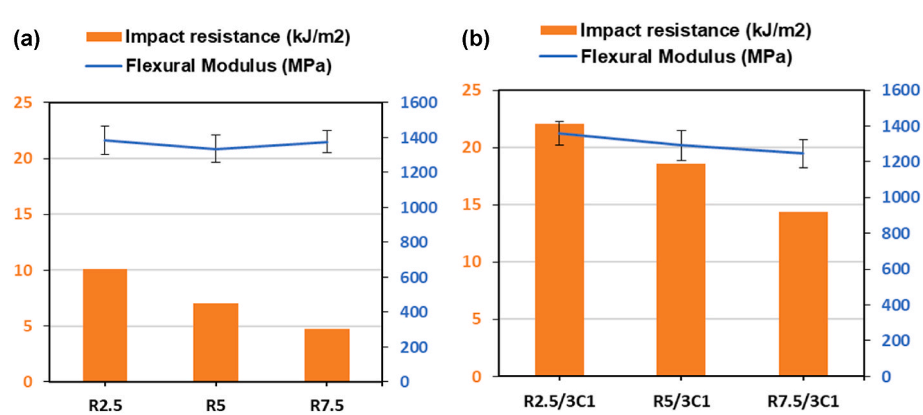

Fig. 4. Mechanical properties of HDPE-PP blends (a) and the same blends with compatibilizer (b).

performance.

For blends between virgin and rHDPE, the results of the physicochemical properties show that both the density and the MFI decrease with the increasing content of the virgin PE, with values between those of the recycled and virgin PE. This behavior is in accordance with the additive principle of the law of mixtures, as the measured property depends on the related properties of the pure components and is related to the volume fractions of the blend (Gooch, 2011). Other properties like the hardness and ESCR are only slightly impacted by the addition of higher contents of virgin resins, which suggests that these properties are less sensitive to the presence of impurities. This is essential for the ESC resistance, as it is a key property for HDPE containers of food and cosmetic products.

Regarding the mechanical properties of these materials, two crucial properties must be considered in the design of HDPE bottles, which are the rigidity of the bottle (flexural modulus) and its robustness (impact strength resistance). Analyzing the values obtained for the flexural modulus (Fig. 5-left), this property is slightly affected by the addition of the virgin resin, independently of the content of virgin PE, probably because the addition of even moderate amounts of rPE causes the flexural modulus to stabilize around a fixed value. On the other hand, the impact resistance increases with the addition of higher amounts of virgin HDPE (Fig. 5-left). However, this augment is not linear, as it only changes from 3 kJ/m^2 to 4 kJ/m^2 at 25 wt% of PE and adding up to 75 wt% PE only increases the resistance to a value of 8 kJ/m^2. The presence of PP in the recycled resin, as confirmed in section 3.1, appears to be a limiting factor, and the addition of only virgin resin does not seem to solve this issue, as all impact resistance values are below the references for this application. Thus, this result leads to the search for other alternatives to help homogenize the PP present in the post-consumer bottle. Therefore, as it has been previously described, a compatibilizer could help to overcome this problem. Thus, an amount of 3 wt% C1 compatibilizer was added to each blend system previously prepared. Properties of each sample are condensed in Table 2. For these blends, the compatibilizer's presence in the polymer matrix has a negligible impact on both the density and the MFI, with a minimum decline in density when compared to the system without the compatibilizer. Other important properties like hardness or ESC resistance follow the same trend as the previous results when only virgin resin is added (Fig. 5-right), indicating that the structural properties of the bottle are not compromised by the addition of the C1 additive to the blend.

Then, flexural modulus and impact strength resistance were evaluated. In this system, the addition of the compatibilizer causes a decrease in the flexural modulus respect the previous results adding only virgin resin, between 100 and 200 MPa. This could limit the stiffness required for the bottle, and therefore, the addition of compatibilizers should be done with caution. Nonetheless, the impact strength resistance its greatly increased with the addition of C1. Although the addition of 25 wt % PE is not sufficient to reach the reference values, the addition of 50 wt % and 75 wt% satisfies the minimum requirements, which are within the estimated range for blow moulding grades and therefore meet the mechanical specifications required for such products (Table S4 in Supplementary Information summarizes the reference values from different companies commonly used to manufacture blow moulding HDPE grades). These results prove that the use of compatibilizing agents could be a solution to improve the properties of milk bottle recyclates contaminated with other polymers, although this addition must be controlled to guarantee that critical properties are not greatly affected, thus compromising the final properties of the material and therefore the final product.

The economic factor must also be considered when adding compatibilizing agents, as costs could rise rapidly, making the use of recyclates less attractive to industry. However, the present and future obligation to incorporate certain amounts of recycled material in newly manufactured products should be high on the agenda of plastics manufacturers.

Additionally, to assess the effect of the addition of a compatibilizing agent during blend preparation, SEM microscopy was performed. SEM micrographs of the fracture surface of impact strength samples for blends containing 25 wt% of virgin PE resin and different contents of C1 compatibilizing agent (1, 2 and 3 wt%) are shown in Fig. 6. The fracture surface of the blend containing 25 wt% of virgin resins (a) shows a smooth surface, characteristic of a brittle behaviour and therefore very low impact resistance. The addition of 1 wt% of compatibilizer hardly shows changes in the fracture morphology of the sample (b). However, micrographs show that the addition of 2 wt% (c) and 3 wt% (d) of C1 compatibilizing agent results in the development of wider shear lips

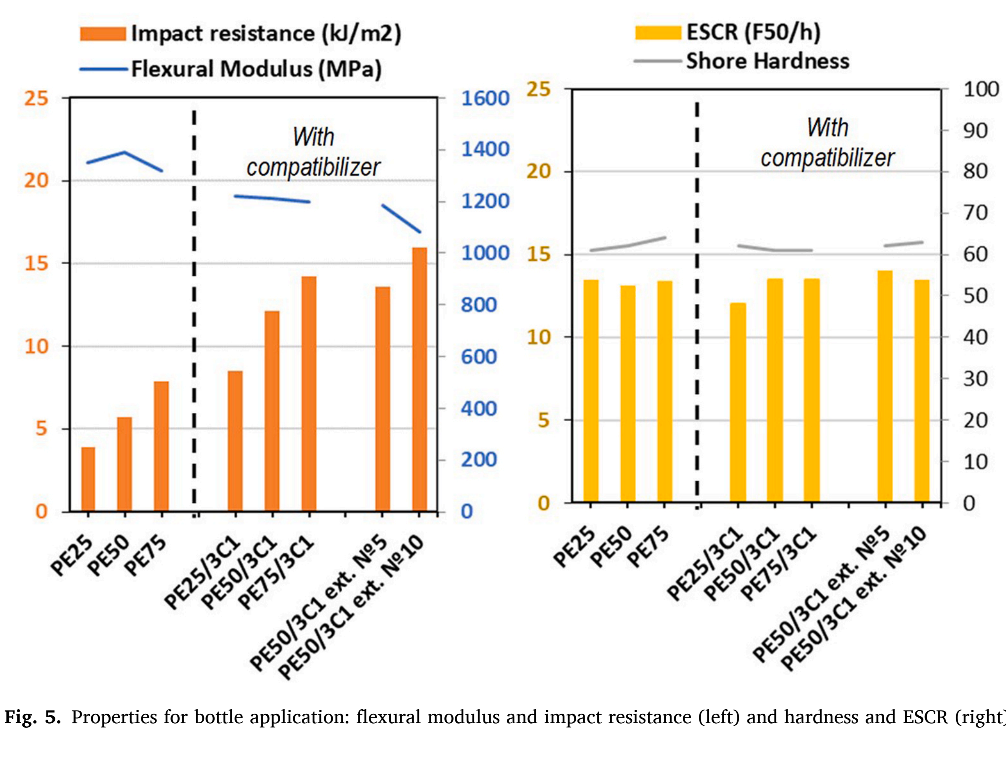

Fig. 5. Properties for bottle application: flexural modulus and impact resistance (left) and hardness and ESCR (right).

Table 2
Physicochemical properties of virgin and recycled HDPE blends with and without compatibilizer and after successive extrusion cycles (5x and 10x).

Blend	Content	Density (kg/m^3)	Melt Index (g/10 min)	Flexural Modulus (MPa)	Impact Resistance (kJ/m^2)	Hardness (Shore D)	ESCR (h)
PE25	25%PE+75%rPE	978	0.69	1351	3.9	61	13.5
PE50	50%PE+50%rPE	970	0.48	1389	5.7	62	13.1
PE75	75%PE+25%rPE	968	0.34	1319	7.9	64	13.4
PE25/3C1	25%PE+75%rPE + 3% C1	972	0.51	1221	8.5	62	12
PE50/3C1	50%PE+50%rPE + 3% C1	967	0.43	1212	12.1	61	13.5
PE75/3C1	75%PE+25%rPE + 3% C1	963	0.34	1197	14.2	61	13.5
PE50/3C1 5x extruded	50%PE+50%rPE + 3% C1	969	0.43	1186	13.6	62	14
PE50/3C1 10x extruded	50%PE+50%rPE + 3% C1	967	0.34	1081	16.0	63	13

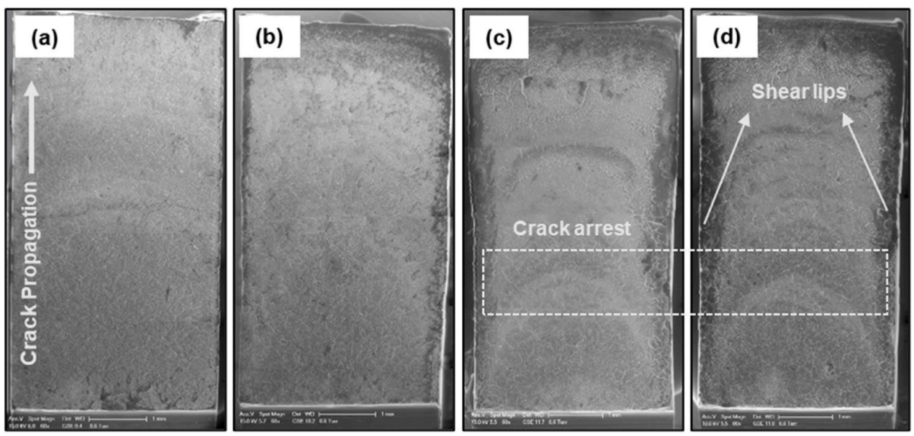

Fig. 6. SEM micrographs of the fracture surface for blends containing 25 wt% of virgin resin (a), and 1% (b), 2% (c) and 3% (d) of compatibilizing agent C1.

with thicker arrest lines indicative of a more ductile failure morphology. These results agree with the increase in impact strength observed when higher amounts of compatibilizer are used during blending.

In a circular economy, it is important to consider not only the use of recycled materials but also what may happen to the recycled materials in subsequent recycling processes, as additives and other substances

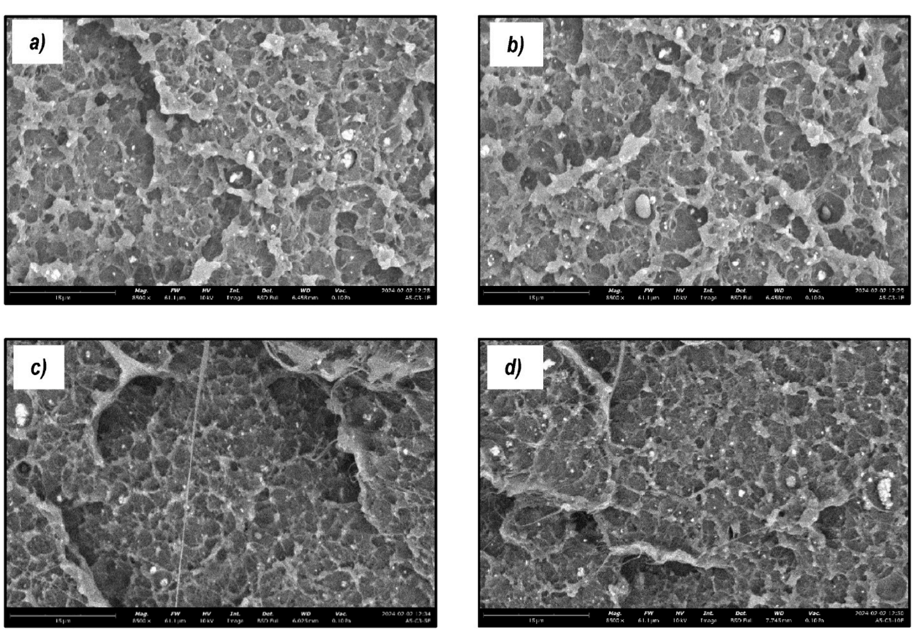

Fig. 7. SEM micrographs of the fracture surface near the notch for blends containing 50 wt% of virgin resin and 3% of C1 (a), and several extrusion cycles: 1 cycle (b), 5 cycle (c) and 10 cycle (d).

present in the recycled material could affect the resulting properties of the blends (Maris et al., 2018). Therefore, several extrusion cycles (5x and 10x extrusion cycles) were simulated on the blend containing 50% of rPE and 3% of compatibilizing agent C1 to assess the potential degradation of the material and how its mechanical properties might be affected in the short- and long-term.

After analysing the results, it is clear that there are no significant changes in the properties evaluated for the blend between successive extrusion cycles, including the 5x and 10x extrusion cycles, in line with conclusions reported by other studies (Boldizar et al., 2000; Gaduan et al., 2023a; Apone et al., 2003). The density remains constant, although a slight decrease is noted in the MFI after the 10x extrusion cycle, suggesting a branching process during the consecutive extrusion cycles (Oblak et al., 2015; Kealy, 2009). There is also a slight decrease in flexural modulus, which could again be attributed to polymer chain thermal degradation caused by extrusion processes (Gaduan et al., 2023b). However, two important aspects can be noted: firstly, impact resistance remains unchanged and even increases slightly; and secondly, environmental stress cracking resistance (ESCR) remains stable, preserving the long-term strength of the material (Fig. 5).

SEM images of the fracture surface of Charpy impact specimens were taken for all blends at different extrusion cycles (Fig. 7). The images taken near the notch reveal that the matrix fibrillation was thick and short in all specimens, with no domains or significant degradation in the polymer matrix. This confirms the good resistance of the blend to subsequent extrusion cycles.

Thus, it could be concluded that with the addition of 50% of virgin resin and 3% of compatibilizer the overall properties of the material are satisfactory, especially the critical ones as the flexural modulus, impact strength resistance and ESCR. Also, the successive extrusion cycles reveal little decrease on the main properties of the material, which is a good indicator of the resistance of the blend. These results demonstrate the potential of incorporating recycling material from milk bottles in the production of HDPE bottles, with a circular economy of open-loop recycling in consideration. However, the presence of other polymers, mainly PP, must be considered, especially in view of the application of the new European regulation that requires the cap and bottle to be kept joined together. In this case, the use of additives is critical to increase the properties of the blends.

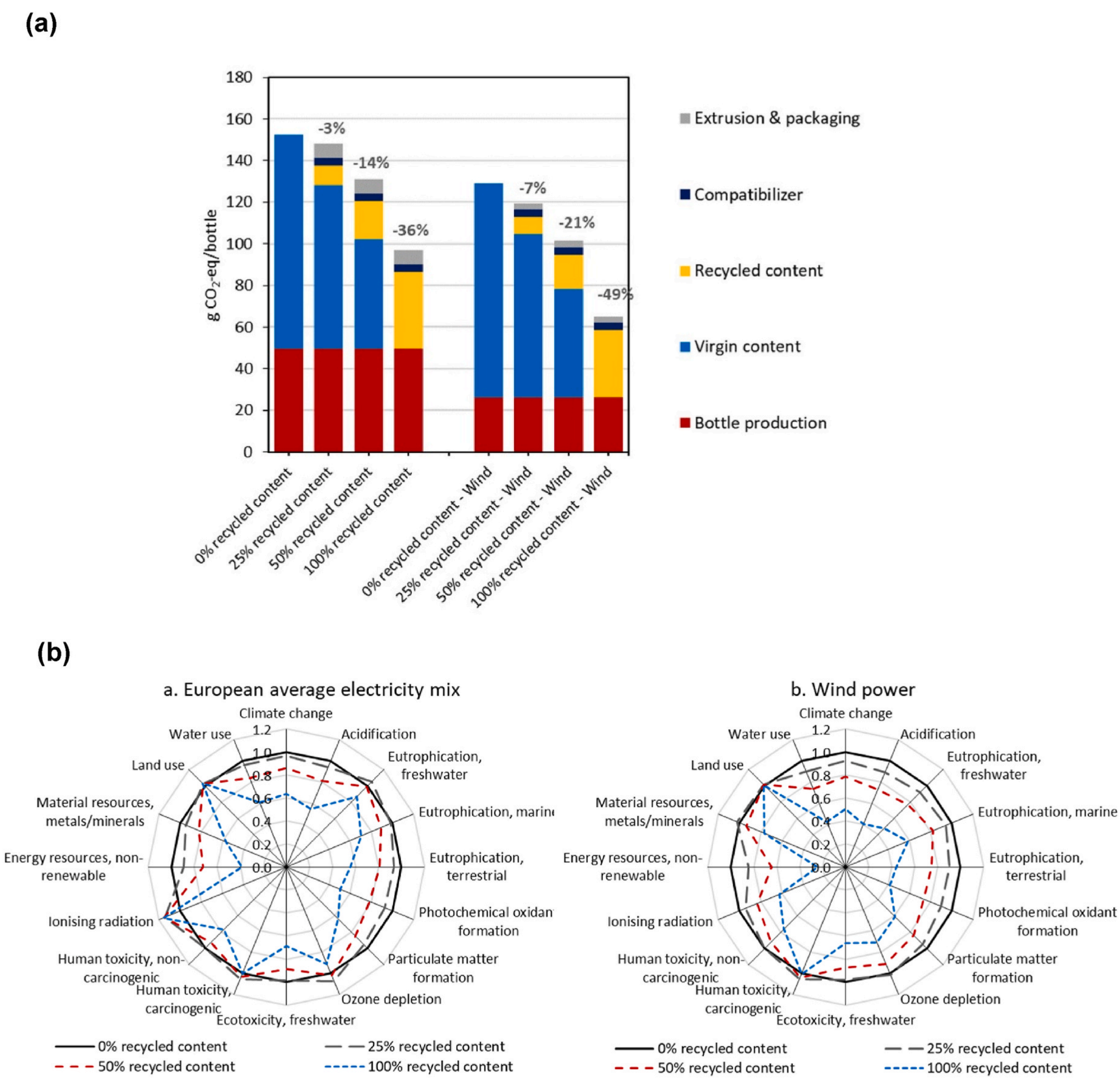

Fig. 8. Carbon footprint results for the production of a HDPE bottle incorporating recycled material and its comparison with a 100% virgin HDPE bottle (a) and relative environmental life cycle impacts of the production of a HDPE bottle incorporating recycled material with respect to a 100% virgin HDPE bottle (b). Impacts were calculated considering two types of sources for powering the supply chain: the European average electricity mix and wind electricity.

3.4.2. Environmental characterization of HDPE bottles with recycling material

Fig. 8a presents the carbon footprint of the production of a non-food HDPE bottle according to the scenarios described earlier. The results show that compared to a 100% virgin bottle (i.e., 0% recycled content), incorporating 25% recycled material leads to a 3% reduction in carbon footprint, while incorporating 50% recycled material results in a 14% reduction. For a bottle with a 100% recycled content, a reduction of 36% is achieved. The reduction is rather modest, particularly observed in the case of 25% recycled content, because a high share of the total carbon footprint is found in the production of the virgin content as well as the bottle production stage, which is not affected by the incorporation of the recycled plastic. Within this context, further reductions could be achieved by switching to renewable energy. Notably, when considering the use of wind power throughout the bottle supply chain, the reductions are slightly higher: 7% (25% recycled content), 21% (50% recycled content), and 49% (100% recycled content), respectively.

Fig. 8b shows the LCA results for a complete set of impact categories obtained using the EF method (Fazio et al., 2018). Generally, similar trends as for the carbon footprint were found for the other categories. Important impact reductions are observed for acidification, eutrophication, photochemical oxidant formation, freshwater ecotoxicity, non-renewable energy resources, and water use. The scenario with 100% recycled material and wind power exhibits the highest reductions across all the categories but metals and mineral resources as well as land use. Overall, despite the limitation on the maximum recycled content, these results demonstrate that environmental benefits can be achieved, especially if advancements in recycling technologies and regulatory conditions allow for a bottle made of 100% recycled material from bottle-to-bottle open-loop processes.

These findings reveal the importance of systematic evaluation in analysing the benefits of circularity and efficient material design for packaging (Pomponi et al., 2022). As the use of recycled material continues to increase, it becomes imperative for the packaging industry to systematically evaluating the environmental impacts of recycled material incorporation, facilitating continuous improvement in sustainability practices. This should be accompanied by strategies that monitoring and responding to potential changes in markets and supply chains. The integration of these aspects into decision-making processes may allow packaging manufacturers to identify opportunities for innovation and optimization, paving the way for a more sustainable packaging industry.

4. Conclusions

Plastic pollution and global concerning regarding this issue has led to an increase in new legislations and policies. While the recycling of plastics has been on the rise last years, many recycled plastics are still not used due to poor properties or lack of real applications. One of these cases is the HDPE milk bottles, as the difference in properties with other HDPE containers such as home and personal care, and the presence of additives like carbon black in some of its layers has caused that milk bottles are currently removed during mechanical recycling to prevent contamination, resulting in a low-value plastic.

This study has focused on finding opportunities for recyclates obtained from HDPE milk bottles in order to promote the circular economy for this particular plastic waste flow. Therefore, its incorporation in the manufacture of HDPE bottles has been explored. The characterisation of this recyclate has revealed the presence of other substances in the polymeric matrix, mainly PP, which due to their incompatibility, cause a deterioration in the mechanical properties of the recycled milk bottles. This fact limits their potential recyclability, which represents a challenge to be solved in the context of mechanical recycling, which is very useful and used in the recycling of mono-plastics but less so in the recycling of mixed plastics.

To solve this issue, different compatibilizing agents have been tested, as well as the addition of virgin resin to improve the properties of the recyclate, both in different amounts to analyse its effect. Although the addition of PE was not sufficient to meet the minimum requirements for the intended application, the addition of a compatibilizing agent increased the impact strength of the blends without a significant lowering of the flexural modulus. In this context, three possible scenarios of contamination have been studied, considering that the trend, due to European legislation, is that the cap (PP) will be attached to the bottle in the very near future and that, therefore, PP contamination is likely to be higher than at present and, moreover, unavoidable in the current mechanical recycling processes. The results obtained show that a maximum of 50 wt% of recycled post-consumer bottle with a 3 wt% of compatibilizer could be used to obtain a blend that fulfil the required properties of stiffness and robustness of HDPE bottles, and therefore, the post-consumer bottles could be reused helping to establish a Circular Economy in this sector. This is further emphasized by the degradation study conducted through simulating successive extrusion cycles, up to a total of ten. It was observed that the critical properties were minimally affected, thus confirming the favourable characteristics of the blend obtained when incorporating this recyclate. Moreover, incorporating 50 wt% recycled material into the bottle would lead to a 14% reduction in the carbon footprint. A larger reduction (up to 49%) could be achieved if combining a bottle of 100 wt% recycled material with a supply chain powered by wind electricity.

Although further studies are needed to address the possible migration of contaminants in these recycled resins to obtain their approval for food-contact applications, in the meantime, the mechanical performance of the blends obtained demonstrates that they could still be used in the non-food packaging plastic industry. In addition, improved ecodesign of packaging and improved sorting, separation, and mechanical recycling processes of HDPE milk bottles may result in higher-quality recycled resins, thus reducing the need for virgin PE and opening new possibilities for plastic waste.

CRediT authorship contribution statement

Aymara Blanco: Writing – original draft, Investigation. **Rafael Juan:** Writing – review & editing, Writing – original draft, Validation, Investigation. **Robert Istrate:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Conceptualization. **Beatriz Paredes:** Writing – review & editing, Supervision, Conceptualization. **Mario Martin-Gamboa:** Writing – review & editing, Writing – original draft, Validation, Conceptualization. **Carlos Domínguez:** Writing – review & editing, Supervision, Methodology, Funding acquisition. **Javier Dufour:** Writing – review & editing, Visualization, Validation, Methodology, Conceptualization. **Rafael A. García-Muñoz:** Writing – review & editing, Visualization, Validation, Supervision, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cesys.2024.100185.

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