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Quality evaluation and economic assessment of an improved mechanical recycling process for post-consumer flexible plastics

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ABSTRACT

Packaging represents the largest fraction of plastic waste in Europe. Currently, mechanical recycling schemes are mainly focused on the recovery of rigid packaging (like bottles), while for flexible packaging, also called films, recycling rates remain very low. Existing mechanical recycling technologies for these films are quite basic, especially in the case of complicated post-consumer flexible plastics (PCFP) waste, leading to regranulate qualities that are often subpar for renewed use in demanding film applications. In this study, the technical and economic value of an improved mechanical recycling process (additional sorting, hot washing, and improved extrusion) of PCFPs is investigated. The quality of the four types of resulting regranulates is evaluated for film and injection molding applications. The obtained Polyethylene-rich regranulates in blown films offer more flexibility (45–60%), higher ductility (27–55%), and enhanced tensile strength (5–51%), compared to the conventional mechanical recycling process. Likewise, for injection molded samples, they exhibit more flexibility (19–49%), enhanced ductility (7 to 20 times), and higher impact strength (1.8 to 3.8 times). An economic assessment is made between the obtained increased market value and the capital investment required. It is shown that the economic value can be increased by 5–38% through this improved recycling process. Overall, the study shows that it is possible to increase the mechanical recycling quality of PCFP in an economically viable way, thus opening the way for new application routes and overall increased recycling rates.

1. Introduction

Packaging is the main destination for plastics, as 40 % of the total plastic use in Europe (i.e. almost 20 million tonnes) is attributed to this sector (PlasticsEurope, 2021). The share of plastic films which are annually placed in the European market is around 13 to 15 million tonnes, of which nearly 9 million tonnes are used in the packaging sector (Eunomia, 2020) as so-called flexibles. To mitigate the concerns regarding the end-of-life scenarios for plastics, society looks toward moving away from a linear economy based on landfill and incineration as waste management strategies (Hou et al., 2018). Yet, for flexible packaging this transition to a circular economy is challenging mainly due to its complex composition.

Mechanical recycling, as a circular approach towards the management of plastic wastes, remains the dominant commercial technology in plastics recycling, with often proven positive total life cycle impacts (Ferdous et al., 2021). After a first sorting, typically performed in material recovery facilities (MRF) dealing with a broad mix of packaging, current-day mechanical recycling for PCFPs consists of cold washing and regranulation via extrusion (Larrain et al., 2021; Luijsterburg and Goossens, 2014), as illustrated in the bottom line of Fig. 1. However, mechanical recycling for the recovery of flexible plastics is not straightforward (Huysveld et al., 2019; Lazarevic et al., 2010).

There are several reasons for the low recycling rates of PCFP (Bening et al., 2021). Annually, almost 2 million tonnes of films in Europe are produced in multi-layered structures (Kaiser et al., 2018). In these structures, the main polymer is cross contaminated with other polymers or even other materials (Häsänen, 2016; Pivnenko et al., 2015; Ragaert

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et al., 2017; Roosen et al., 2020; Tartakowski, 2010). Furthermore, since films are thin items, their surface-to-weight ratio is also high. Thus, higher levels of surface contamination (like food remaining) are common, as well. Moreover, conventional near infrared (NIR) sorting lines are equipped with nozzles using compressed air to eject the target streams, which is more challenging in case of lightweight mixed flexibles (Jansen et al., 2015; Kaiser et al., 2018).

Additionally, during the pretreatment of the waste, the efficiency of the washing procedures for flexibles is lower, compared to rigids (Niaounakis, 2020). One of the reasons is the fact that rigids are geometrically more stable and do not fold together in the washing process. Hence, the imposed shear by the washing medium will be applied to the interface of the shredded plastic flake and any residue stuck to it rather than being relaxed by a change in the geometry of the flake (Niaounakis, 2020). On top of this, the current washing processes fail to remove components such as inks, different polymer layers, non-polymer layers (like aluminum and paper), etc. (Roosen et al., 2022). Thus, these components will end up in the extrusion process, together with the main polymer (Roosen et al., 2020; Ügdüler et al., 2021).

The pretreated plastic flakes then go to the next step of the recycling process, i.e. regranulation, where the plastic flakes are melt-mixed and pelletized into regranulates. However, conventional "general" industrial extrusion lines are not equipped with sufficient filtering and degassing modules (Luijsterburg and Goossens, 2014), required for the processing of highly contaminated fractions like recovered PCFPs, limiting the processability of the fraction, and yielding a regranulate of low physical and aesthetical quality, which can only be downcycled to few applications.

To obtain higher recycling rates, next to the technical challenges, there are several economic concerns as well. To date, the technology and economic assessments of packaging waste treatment mainly focus on MRFs (Cimpan et al., 2016; Da Cruz et al., 2014, 2012; Marques et al., 2014) and research on the actual mechanical recycling, especially for PCFP waste, is scarce. The results of these studies on the economic structure for household waste collection and sorting at MRFs indicate that most of the costs should be supported by an Extended Producer Responsibility (EPR) scheme, e.g., gate fees. This demonstrates that the revenues from their sales are lower than the cost incurred by collection and sorting of household packaging waste. One of the costs related to recycling of Polyethylene (PE) films is the price of the feedstock that can range from positive to negative values depending on the quality of the bales created at MRFs (Cimpan et al., 2016; Larrain et al., 2021). The current price of regranulates is significantly lower for films compared to other rigid polymers, mainly due to aforementioned high contamination of bales. Literature highlights the need to increase high quality regranulates production from the collected PCFP (Brouwer et al., 2018; Faraca and Astrup, 2019; Ragaert et al., 2017). In this context, an improved mechanical recycling process is proposed by the industrial CEFLEX consortium, called the Quality Recycling Process (QRP), which is based on existing technology, but with more advanced sorting and recycling steps (Mosora, 2020).

QRP focuses on the two main bales which are recovered from PCFPs, i.e. DSD 310–1 and DSD 323–2 (Der Grüne Punkt, 2021). DSD is the abbreviation for 'Duales System Deutschland (DSD) GmbH', which is the German dual system and has developed the standards for the degree of allowed contamination in the sorted bales. DSD 310–1 is composed of at least 92 wt% plastic films, with dimensions larger than an A4 paper, which are mostly made of PE. DSD 323–2 consists of at least 90 wt% plastic films, which are mostly made of either PE or Polypropylene (PP), i.e. it has a polyolefin (PO) composition.

QRP, which is evaluated at semi-industrial scale in the current study, involves several instrumental and technical improvements at different stages compared to the conventional mechanical recycling process, which are schematically illustrated in Fig. 1. Within this new approach, to reduce the cross contaminations of polymers, the bales are further sorted into more pure bales with an additional set of optical NIR sorters (Mosora, 2020). From DSD 310–1, transparent clear PE films are separated as a new bale, which is called "PE Film Natural" in this study. The other bale which is separated from DSD 310–1 is called "PE Flex", as this is still mainly flexible PEs but of different colors. However more polymeric cross contaminations are present compared to PE Film Natural. From DSD 323–2, PP film (all colors) is separated and called "PP Film". The other bale is called "PO New". The latter is a blend of PP and PE, but with lower PE content compared to PE Flex. Fig. 2 depicts the flow of materials in QRP and also the composition of each regranulate after different recycling processes. More detailed information regarding the configurations within the sorting equipment and also the mass balances are presented by Lase et al. (2022).

After sorting, the bales are transported to a recycling facility for washing and regranulation. To improve cleaning, QRP Tier 1 approach proposes a hot washing process (>80 °C) with detergents and caustic soda on top of the cold wash of the conventional recycling (Mosora, 2020). Prior to washing, the films are shredded, using a 30 mm sieve, and within the washing process, the sunk fraction is excluded as it is mostly of non-PO constituents observing a density larger than that of the washing medium. Moreover, in QRP Tier 1, the washed flakes are regranulated in extruders equipped with an extra 125 μm melt filter next to the single step filtration (90–110 μm) of the conventional recycling. Previous research showed that the odor of post-consumer recycled plastics is another aspect limiting their applicability (Demets et al., 2020; Strangl et al., 2020). Therefore, in QRP Tier 1, after the extrusion, the regranulates are deodorized in dedicated equipment (Mosora, 2020) based on hot gas flows. In parallel, in the Tier 2 procedure, the additional sorting is followed by the conventional recycling process, i.e. the bales are cold washed and a single step filtration in extrusion is used and there is no deodorization (illustrated in middle line of Fig. 1). Obviously, whereas Tier 2 is a less expensive process, it is expected to yield lower purities. More details regarding the process and a material flow analysis

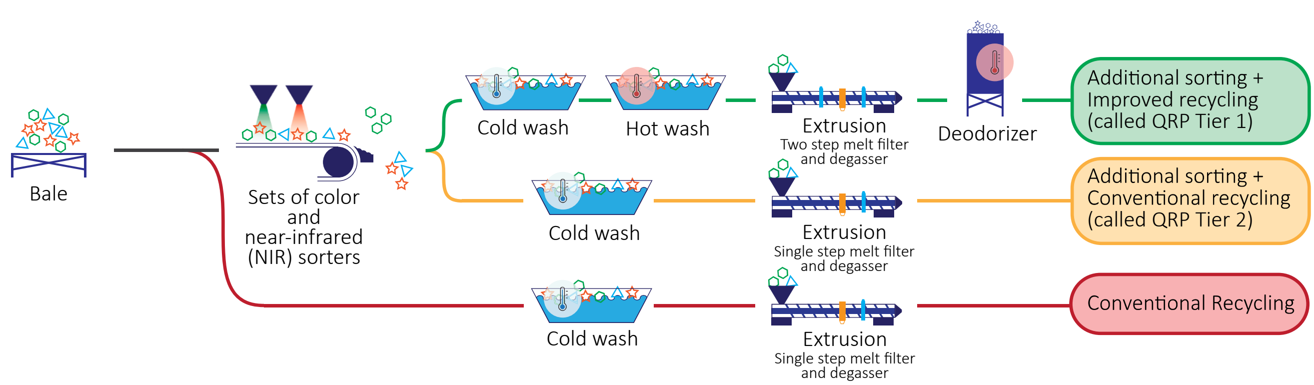

Fig. 1. Scheme of a current conventional mechanical recycling process (bottom), which can be expanded either with only additional sorting (middle) or replaced entirely by additional sorting and improved recycling (top).

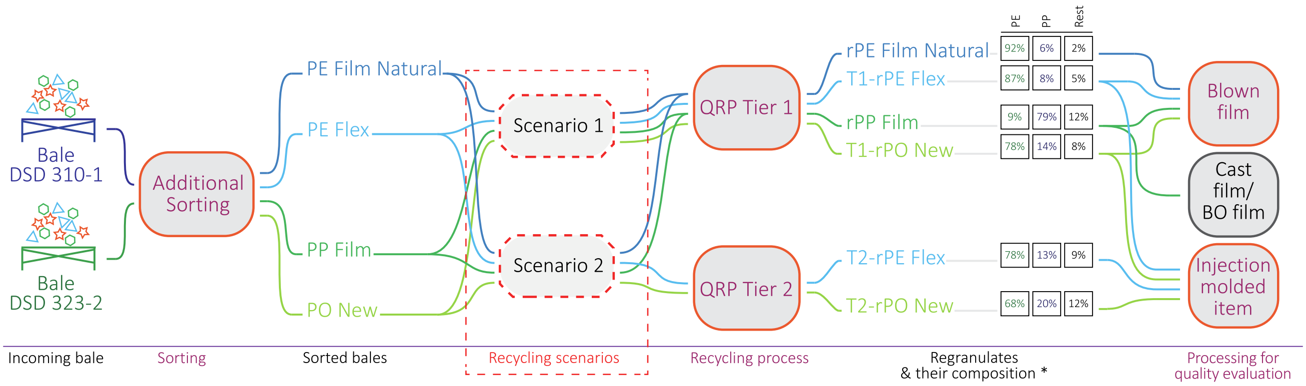

Fig. 2. Flow of materials within different steps of QRP at two scenarios and the composition of the regranulates (*= compositional data are taken from Lase et al., (2022)). Cast film and biaxially oriented (BO) film samples are produced and provided by external partners.

can be found in Lase et al. (2022). In summary, the technical improvements of QRP include (i) additional sorting, (ii) hot washing with detergent, (iii) improved extrusion, and (iv) deodorization.

Previous studies reporting on PCFPs are rare. Those which exist are typically limited to either an analysis of the bale composition (Brouwer et al., 2018), high-level techno-economic analysis (Larrain et al., 2021), life cycle analysis (Horodytska et al., 2020; Hou et al., 2018; Martín-Lara et al., 2022) or to a very specific aspect like delamination (Ügdüler et al., 2021). Manuscripts which do explore the properties of resulting regranulates (and which products can be made from them), typically do this at lab-scale only (Huysveld et al., 2022).

Hence, the current research is the first study on PCFP, which (i) is done at a representative industrial scale, (ii) includes the properties of the final regranulates and (iii) includes validation with effectively manufactured commercial products. This research aims to open the way for further explorations towards the advancement of the existing mechanical recycling procedures for PCFPs. It assesses the technical and economic value of QRP compared to both the conventional recycling process and to reference virgin grades. The quality of regranulates is evaluated for film and injection molding applications. Finally, an economic assessment is made between the obtained increased market value and the required capital investment.

2. Materials and methods

2.1. Materials

Two commercially pre-sorted PCFP streams are sourced as incoming bales DSD 310–1 and DSD 323–2 and ran through pilot sorting and recycling lines of QRP. The trial size at input level is 3.6 tonnes for DSD 310-1 bale and 3.5 tonnes for DSD 323-2 bale. Fig. 2 and Table 1 summarize the origin and composition of the studied regranulates within this paper. More details can be found in Lase et al. (2022). The PE Film Natural and PP Film bales are always processed according to Tier 1. The PE Flex and PO New bales are tested both in Tier 1 (T1) and Tier 2 (T2). The nomenclature is given accordingly in Table 1. Once a material has gone from sorted bale to extruded regranulate, an 'r' is added to the nomenclature, e.g. the sorted bale PP Film becomes the regranulate rPP Film. The regranulates of the materials are dried at 60 °C for 48 h in a vacuum oven to minimize the moisture content, prior to characterization and melt processing.

2.2. Physical properties of the regranulates

Density measurements are performed according to ISO 1183 on granules and also on injection molded samples. Bulk density of the regranulates is also measured according to ISO 60. Reported values are an average of five measurements.

The melt flow index (MFI) is measured according to ISO 1133 using a Tinius Olsen MP1200 at both 190 °C and 230 °C, each time for a weight of 2.16, as well as 5 kg. Reported values are an average of six measurements.

2.3. Quality evaluation of the regranulates

As has been elaborated by Demets et al. (2021), the concept of quality of a regranulate is tied to the intended application (which determines the method of manufacture), both in terms of which properties are considered essential, in terms of whether their values are expected to be high or low and how they are measured. For example, in films, impact strength is rated via a dart drop test, while in injection molded goods, it is evaluated via a Charpy impact test. Likewise, flexible blown films require low E modulus (also called elastic modulus or Young's modulus) values, while components for electronics often require high rigidity.

Therefore, as is illustrated in Fig. 2, specimens are produced in different selected processes (film blowing, cast film extrusion or injection molding) and are tested to determine the key properties for these type of applications, which are listed in Table 2. The E modulus is a material's resistance to elastic deformation and in fact describes its initial response to being loaded. As such, it is the dominant property on which the mechanical functionality of materials is compared (Ragaert et al., 2020b). A close second in importance is the tensile strength, which is a measure for how much stress a material can tolerate prior to failure. Finally, the strain at break is used to evaluate how much deformation a material can exhibit until failure. These three properties together give a complete image of how a material (and the products they are made of)

Table 1
Recycling conditions for the studied regranulates, sourced from DSD 310–1 and DSD 323–2 bales.

Bale name	Washing condition	Extrusion melt filters	Deodorization	Regranulate name	Recycling scenario
DSD 310–1	Cold & Hot	Two step	Yes	rPE Film Natural	1 & 2
	Cold & Hot	Two step	Yes	T1-rPE Flex	1
	Cold	Single step	No	T2-rPE Flex	2
DSD 323–2	Cold & Hot	Two step	Yes	rPP Film	1 & 2
	Cold & Hot	Two step	Yes	T1-rPO New	1
	Cold	Single step	No	T2-rPO New	2

Table 2
Key properties for the selected applications.

Specimen production method	Key properties
Blown film	Elastic Modulus, Tensile Strength, Strain at Break, Dart Drop Resistance
Cast film	Elastic Modulus, Tensile Strength, Strain at Break
Biaxially oriented film	Elastic Modulus, Tensile Strength, Strain at Break, Thermal Shrinkage
Injection molded items	Elastic Modulus, Tensile Strength, Strain at Break, (Notched Charpy) Impact Strength

will behave under quasi-static loading. Therefore, they are considered key properties for all investigated applications. For the injection molding and blown film applications, impact resistance is also important. They are measured by the so-called dart drop test for films and the notched Charpy impact test for injection molded goods. Finally, thermal shrinkage in biaxially oriented films is an important property to guarantee their suitability for certain applications. The detailed information regarding the preparation of specimens and their mechanical testing is included in the Supporting Information (SI).

The QRP regranulates' experimentally determined values of these properties are then scaled in a comparative diagram. The properties of the current baseline (the whole incoming bale without additional sorting, processed conventionally) are set as the '1′ values and all other materials have their values recalculated proportionally. This allows for a comparison of the obtained regranulates with the performance of both the current baseline, an equivalent virgin, and a post-commercial and industrial (C&I) recycled PE. C&I regranulates are sourced in large quantities from enterprises which have production scraps or discarded plastics in form of like temporary packaging before the assembly of the content. These materials are mostly of higher quality compared to post-consumer regranulates, as they are more uniform in composition and less contaminated.

The absolute reference values and their sources are included in Table SI1 of the Supporting Information. It is noteworthy that the reference materials are carefully selected as belonging to the relevant application category. For film, only the Tier 1 materials are considered, as this process is more sensitive to contaminants. For injection molding, the Tier 2 materials are also considered. Based on the performance of the new regranulates in the comparative diagrams, the suitability (or lack thereof) of each regranulate for an end application is determined and industrial demonstrators are selected for production trials.

2.4. Economic assessment

In the economic assessment, a cost-benefit analysis (CBA) is performed of the recycling process by quantifying the difference between the processing costs incurred and the revenue generated from regranulates sales. The economic balance is given without external financial support, which are normally paid by the Producer Responsibility Organization (PRO) (Da Cruz et al., 2014, 2012; Marques et al., 2014). The capital investment (CAPEX) and annual operational costs (OPEX) associated with the recycling processes are estimated by collecting the required data points from previous studies (Cimpan et al., 2016; Larrain et al., 2021), machine builders' specifications, and expert judgment, the data points from literature are used when the information suit the investigated (improved) mechanical recycling processes in this research (e.g., machines' processing capacity), followed by consultation with the machine builders and experts from the recycling industry (members of the CEFLEX consortium). Particularly, two QRP scenarios, each processing 20,000 tonnes of DSD 310–1 bale and 20,000 tonnes of DSD 323–2 bale, per year, are compared against the baseline scenario of conventional recycling, processing the same input amount, annually. As can be seen in Fig. 2, in scenario 1, all four sorted bales, i.e. PE Film Natural, PP Film, PE Flex, and PO New are processed through Tier 1 process. In Scenario 2 (Fig. 2), only PE Film Natural and PP Film bales are processed through Tier 1, while PE Flex and PO New are processed to Tier 2. Moreover, the individual processes in the QRP and conventional recycling are grouped into a few plant sections as summarized in the SI (Table SI3 and Table SI4).

The approach to estimate the capital investment is adopted from Sinnott and Towler (2019), which is applied in estimating the capital investment to build plastics waste sorting and recycling plant in previous studies (Cimpan et al., 2016; Larrain et al., 2021, 2020). The calculation of the CAPEX includes the equipment prices plus the additional costs associated with investing in the equipment, namely the installation of equipment costs (IC) and engineering and project management costs (EPMC). On top of these additional costs, the investment cost includes the building and construction (BC) of the actual plant itself. The values of these parameters can be found in Table SI4.

The OPEX is quantified by calculating the cost of energy consumption, transport, disposal fee, general expenses, direct production costs (labors, repair, and maintenance), and fixed costs (depreciation and insurance). The value for each cost parameter can be found in Table SI4. The assessment is based on the plant configuration, scale and material flows as presented in Lase et al. (2022). Moreover, the investment is annualized for 6 – 7 years for the processing equipment (i.e., NIR, washing equipment, dryer, extrusion, etc.) and 10 years for the plant itself.

The price of regranulates from the conventional recycling is obtained from Larrain et al. (2021). Because the projection of low density polyethylene (LDPE) and mixed polyolefin (MPO) regranulates can differ depending on the market condition, here in this research the prices of T2-rPE Flex and T2-rPO New (which are comparable to LDPE and MPO regranulates in a previous study from Larrain et al.(2021)) are set to be €400 and €300 per tonne, respectively, on the basis that regranulates quality might improve with deodorization process, allowing the regranulates to be used in more demanding applications. When processed through Tier 1 recycling, the regranulate price of T1-rPE Flex and T1-rPO New is assumed to be higher, i.e. €500 and €400, respectively. Additionally, this improved recycling process creates two more bales (i.e., rPE Film Natural and rPP Film), for which prices are not yet determined in the market for post-consumer regranulates. However, as the technical properties of these regranulates are significantly improved, we assume that the price will get close to the price of virgin plastics. Hence, the price of rPE Film Natural and rPP Film is set to be €1200 and €1300 per tonne respectively (Plastic Portal EU, 2021; Plasticker, 2021).

A sensitivity analysis is performed to examine how cost modeling parameters and the price of regranulates can influence economic balance of QRP. Sensitivity analysis is done by altering each of the selected parameters (electricity cost, depreciation, and labor costs), price of regranulates, and investment of the selected recycling equipment (debaler, shredder, washing equipment, and extruder) individually by $\pm$ 25 % (detailed information is provided in Table SI5).

3. Results and discussion

In this section the physical properties of all regranulates are presented first. Then, the performance of the regranulates as films and injection molded items are systematically discussed. For each of these two processing methods, the processability is assessed (in the SI, Section 2), the mechanical performance is investigated, and the resulting quality evaluation is presented. Every time, this quality evaluation is validated into an industrial demonstration product with Tier 1 regranulates. After this technical assessment, the results of the economic analysis are presented.

3.1. Physical properties of the recycled regranulates

The results of the MFI measurements are shown in Table SI1. The tested materials observe MFI values below 1 dg/min (at 190 °C and 2.16

kg), which is in agreement with the compositions in Fig. 2 as being mostly of low MFI components, namely the PEs, which are used in the initial film blowing applications (Patel, 2016; Viksne and Bledzki, 1998). The exception is rPP Film, which is mostly composed of PP materials which are derived from biaxially oriented PP films (Breil, 2016) and as expected observe higher melt indices compared to the other regranulates.

rPP Film exhibits a MFI value suitable for both extrusion-based and injection molding processes, while the other five materials observe MFI values suitable for mainly extrusion-based applications (Brouwer et al., 2020) like cast film extrusion, profile extrusion or film blowing. Injection molding of these materials might be challenging as their viscosity is not optimal for their flow into potentially thin-walled designs in injection molded items (Yokoi et al., 1994b, 1994a; Yokoi and Han, 2005).

3.2. Film extrusion of all regranulates

3.2.1. Mechanical properties of blown films

The mechanical performance of all four Tier 1 materials (rPE Film Natural, T1-rPE Flex, rPP Film, and T1-rPO New) converted into blown films is summarized in Table 3. According to Lase et al. (2022), rPE Film Natural is composed of over 90 % PEs and rPP Film consists of almost 80 % PP, resulting in stronger and stiffer films. Any further comparison between both regranulates is pointless, given the different material composition (and typical application) of these fractions.

For PE film applications, mostly dart drop resistance is an essential characteristic and film items usually exhibit dart drop resistance values of above 100 g at 60 μm thickness (Kim et al., 1997; Siegmann and Nir, 1987). The deficiency in rPE Film Natural's dart drop resistance can be attributed to the presence of contaminations and phase separation between the PE matrix and the remaining small amounts of PP, both of which can play a significant role, even in low concentrations (Demets et al., 2022; Van Belle et al., 2020). A phase separated polymeric system often exhibits low dynamic mechanical performances (Strapasson et al., 2005; Tai et al., 2000). As such, it is common industrial practice to blend such recycled plastics with virgin or C&I PE to boost the properties (which is not investigated in the current manuscript).

The composition of the rPE Flex and rPO New are more similar to each other (Fig. 2), albeit with a higher PE content (90 %) for rPE Flex than for rPO New (70–80 %). As a result, rPE Flex displays a better mechanical performance compared to rPO New. Both qualities are still very low in dart drop resistance. It is noteworthy that all four regranulates in form of films exhibit very high ductility and deform significantly prior to their rupture.

3.2.2. Quality evaluation for films

Currently the film applications for the DSD 310–1 after conventional recycling are limited to bin bags (Horodytska et al., 2018), in which the regranulate is typically not used as such, but blended in with virgin or C&I recycled PE. The regranulates from the conventional recycling of DSD 323–2 have very limited applications, none of which are in film. The material typically goes to robust profile extrusion for the production of park benches or flooring slabs (Faraca and Astrup, 2019). In the next part, the quality of QRP regranulates is assessed.

3.2.2.1. Quality evaluation for rPE film natural, rPE Flex, and rPO new – blown film. In Fig. 3, the key properties of the rPE Film Natural, rPE Flex, rPO New, a representative virgin film grade PE material, a C&I recycled PE, and the baseline, which is DSD 310–1 upon conventional recycling, are plotted against each other. The baseline value of DSD 310–1 is too stiff (high E modulus), too weak (low tensile strength) and brittle (low dart drop) compared to the virgin LDPE. It can be seen that the properties diamond of the rPE Film Natural is moving more towards the high quality recycled plastics (i.e. C&I materials) and virgin materials. Tensile strength and strain at break of rPE Film Natural are improved each by almost 50 % compared to the baseline, i.e. conventional recycling. The difference of the E modulus of rPE Film Natural compared to the virgin PE is only 27 %, while the baseline differs by 84 %. Dart drop resistance of rPE Film Natural is only slightly improved compared to the baseline, however this remains a limiting property for the applicability of this material. This low dart drop resistance property is mainly due to immiscibility of different PEs and also the other polymers which are present in the composition, although even in low amounts (Rungswang et al., 2019; Tas et al., 2005). The surface defects caused by contaminations also function as stress concentration points and further degrade the dart drop resistance.

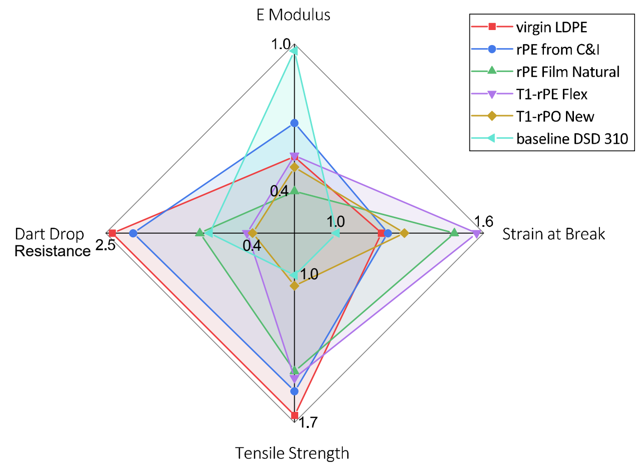

Fig. 3. Mechanical performance comparison between recovered regranulates, baseline and reference materials on blown films. Axes are scaled differently for ease of visual interpretation.

Although being sourced from post-consumer waste, rPE Film Natural observes considerable aesthetical improvements, as shown in Table SI2, and the intensity of the smell in these recovered regranulates are reduced thanks to the deodorization step of Tier 1 process (Strangl et al., 2020). Combining these improvements with aforementioned mechanical performance indicates the potential suitability of rPE Film Natural for a more demanding film application.

An industrial demonstration with this material is run by PepsiCo (The United States), in which 30 % of rPE Film natural is blended into virgin PE for production of collation shrink films at a thickness of 55 μm. Collation shrink film, until now, has been a market into which conventionally recycled DSD 310–1 has been unable to penetrate due to quality issues. The shrink films are further successfully processed and tested for the packaging of six bottles of soda into a sales unit. Some illustrations of the final product are added in the SI (Fig. SI2).

In Fig. 3 it can be seen as well that the film properties of rPE Flex (considering its higher contamination with PP) are also improved

Table 3
Mechanical properties of blown films from rPE Film Natural, rPP Film, T1-rPE Flex, and T1-rPO New.

Material	Thickness (μm)	E Modulus (MPa) - MD	Tensile Strength (MPa) - MD	Strain at Break (%) - MD	Dart Drop Resistance (g)
rPE Film Natural	65 ± 3	129 ± 34	18 ± 4	567 ± 116	57
T1-rPE Flex	60 ± 7	154 ± 30	17 ± 2	455 ± 75	26
rPP Film	58 ± 4	243 ± 91	39 ± 9	973 ± 67	21
T1-rPO New	57 ± 11	163 ± 38	13 ± 2	490 ± 163	<19

towards the virgin film compared to the baseline. The modulus and tensile strength of rPE Flex are respectively equal and only 12 % different from those of the virgin PE. The only property that still lags is the dart drop value, which is a topic for further research. It remains to be seen what properties can be achieved by blending the rPE Flex with virgin or C&I recycled PEs. For film application, the overall performance of rPO New is too low. rPO New has a very low tensile strength, only 5 % different from the baseline of DSD 310–1 and combined with the low dart drop and very limited processing window (as described in SI), this regranulate is clearly unsuitable for film blowing applications. In Section 4.3, both rPE Flex and rPO New regranulates will be investigated for alternative injection molding applications.

3.2.2.2. Quality evaluation for rPP Film – cast films. rPP Film contains almost 80 % PP (Fig. 2). For such a stream, a closed loop recycling for the production of PP films is desirable. Instead of film blowing, PP films are mostly produced by means of casting and further hot stretching which delivers (biaxially) oriented PP morphologies (Breil, 2016). As such, we compare the rPP Film converted to biaxially oriented film (Fig. 4 a) and cast film (Fig. 4 b) to a virgin equivalent for these product types, produced and tested under the same conditions. The absolute values can be found in Table SI1 in the SI. There is no current baseline to compare to, as rPP Film from QRP is in fact a wholly new regranulate stream. The DSD 323–2 in conventional recycling is not suitable for casting processes, due to the non-polyolefin contaminations (Lase et al., 2022).

Fig. 4 shows that in both applications, rPP Film is able to match one key property, while scoring poorly on one other and reaching around 50 % of the virgin value for remaining properties. This indicates that it is unlikely that the material can substitute virgin PP one on one, but the material does merit an exploration of combining it with virgin.

As a demonstration, the rPP Film is successfully processed by Taghleef Industries (Italy) at a 32 wt% rate in a multi-layered biaxially oriented PP (BOPP) structure for a stand-up pouch, wherein the rPP Film forms the inner layer. Illustrations of the final product are included in SI (Fig. SI3).

3.3. Injection molding of the rPE Flex and rPO New regranulates

3.3.1. Mechanical properties of injection molded items

The mechanical performance of the regranulates is tested in uniaxial tension, summarized in Table 4. As rPO New contains higher PP content compared to rPE Flex, it observes a higher E modulus. All materials exhibit tensile strengths in the same range and strain at break values over 100 %, which is considered a good ductility for injection molding products (Van Belle et al., 2020). There is a significant difference in the failure mechanism: both T1-rPE Flex and T2-rPE Flex materials go through a strain hardening with a uniform deformation, while rPO New samples observe a non-uniform deformation. Beyond a certain strain (at around 70 %), necking occurs, and very soon after, the core of the specimen fails. However, the shell of the sample deforms until final fracture. This deformation behavior results in a distinct cup-cone shaped fracture of the rPO New specimens. Samples are collected from the both core and shell of the fractured samples and they are examined with differential scanning calorimetry as described by Lase et al. (2022). No major difference between the composition of the core and shell of the rPO New samples is seen. This difference in mechanical behavior of the core and shell can therefore be attributed to differences in cooling rates through the thickness after injection molding and low interfacial adhesion between the PE and PP phases, causing more massive crystallinities in the core, which are more likely to act as stress concentrators and initiate non-ductile failure (Bajracharya et al., 2016).

Finally, the impact strength of the T1- and T2-rPE Flex is consistently higher than that of rPO New. Somewhat unexpected, T2-rPE Flex appears to be tougher than T1-rPE Flex, as evidenced by both a higher strain at break and impact strength value. There is no immediate explanation for this. While the additional sorting is the same for both trajectories, the Tier 1 materials are expected to contain less non-PO contaminants due to the added filtration step. Fig. 2 shows that T2-rPE Flex contains an estimated 9 % of non-PO, while T1-rPE Flex contains only 4 %. The PP content is also higher, estimated at 13 % for T2-rPE Flex, compared to 8 % in T1-rPE Flex. Potentially, it is this higher PP content that leads to a better strain at break and impact strength of the T2-rPE Flex, which is aligned with the high ductility reported for rPP Film in Table 3 and Fig. 4 b. This notwithstanding, all tested regranulates observe relatively high impact strength values, considering they

Table 4
Properties of injection molded specimens produced from rPE Flex and rPO New regranulates.

Material	E Modulus (MPa)	Tensile Strength (MPa)	Strain at Break (%)	Notched Charpy Impact Strength (kJ/m^2)	Density (kg/m^3)
T1-rPE Flex	335 ± 10	13 ± 1	284 ± 21	(P*) 22 ± 1	950 ± 1
T2-rPE Flex	352 ± 37	13 ± 1	376 ± 8	(P*) 31 ± 4	953 ± 1
T1-rPO New	484 ± 25	14 ± 1	150 ± 49	(C**) 15 ± 1	950 ± 1
T2-rPO New	530 ± 35	14 ± 1	128 ± 52	(C**) 16 ± 1	956 ± 2

*P = Partial break, **C = Complete break.

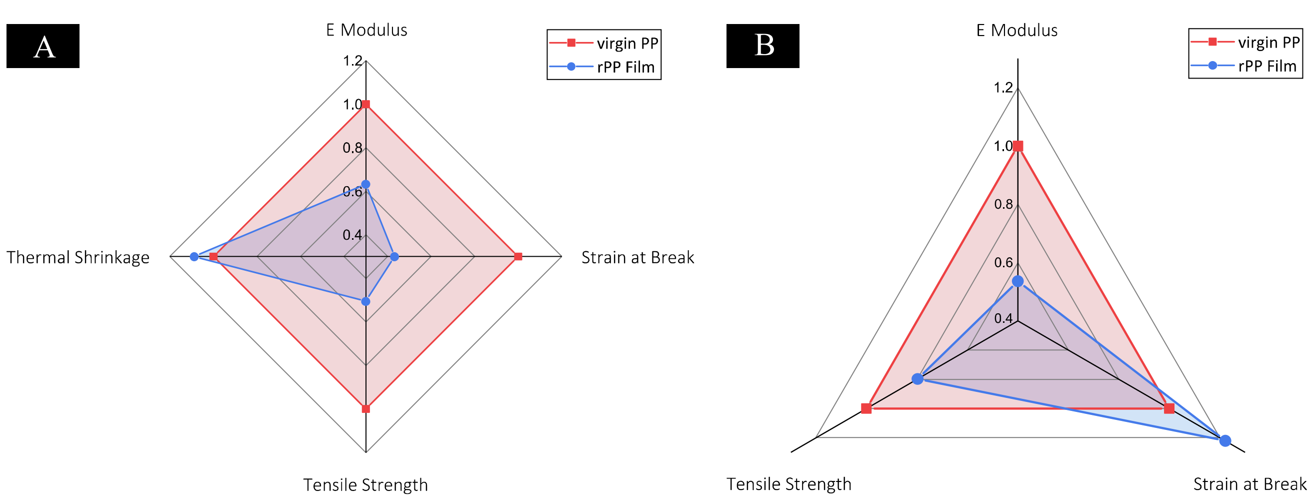

Fig. 4. Mechanical performance comparison between recovered rPP Films and virgin PP on biaxially oriented (A) and cast (B) films.

are mixed polyolefins (Hubo et al., 2016) and therefore this property is not considered a bottleneck for application in injection molding.

3.3.2. Quality evaluation for injected molded items

By comparing the Tier 1 and Tier 2 properties of each regranulate in Table 4, it can be concluded that although hot washing and two-step melt filtration is included in the Tier 1 process, no major change in the performance of the injection molded samples can be obtained. However, the materials from Tier 1 process are lower in contamination and smell (based on our own qualitative impression and supported by Strangl et al. (2020)), giving an advantage to Tier 1 for certain applications. Lower contamination content might deliver measurable improvements on the mechanical performance of the thin walled items, where the impact of non-molten inclusions can be more apparent. Overall, by switching from Tier 2 to Tier 1 only a limited improvement can be achieved, meaning that for these regranulates, i.e. rPE Flex and rPO New, the Tier 2 process should be more economically viable, unless a certain application demands lower smell and inclusion content. The relatively low MFI remains a point of attention, as this may be a practical objection for either thin-walled items, high-speed production and overall energy consumption (higher pressures or temperatures required). Naturally, additives exist that will improve the material flow (Kulikov et al., 2009).

In Fig. 5, the injection molded properties of T1-rPE Flex and T1-rPO New are plotted against a baseline of post-consumer MPO, which is typically applied in very robust products only. While the authors are confident that rPE Flex and rPO New would perform very well in these robust applications (typically by intrusion), it is the goal to validate these materials in more challenging (meaning thinner-walled, with higher demands) applications like injection molding products. Therefore, values are also added for injection molding grades of both a virgin high density polyethylene (HDPE) (rigid applications) and LDPE (flexible applications). The absolute values can be found in Table SI1 in the SI.

The baseline MPO material has a very low impact strength and elongation at break. This overall brittle behavior is in fact the main limiting factor for injection molding applications (Demets et al., 2022). Both T1-rPE Flex and T1-rPO New show significant improvements in strain at break, even if they cannot score as good as the virgin materials. These two materials observe as well higher impact strength values than that of the baseline MPO and virgin HDPE, increasing the application opportunities for injection molding. Of the two materials, T1-rPE Flex is clearly more suited to applications otherwise considered for flexible LDPE, such as flexible closures. Meanwhile, the higher rigidity of T1-rPO New excludes it from flexible applications but it might be suitable for products otherwise made in HDPE.

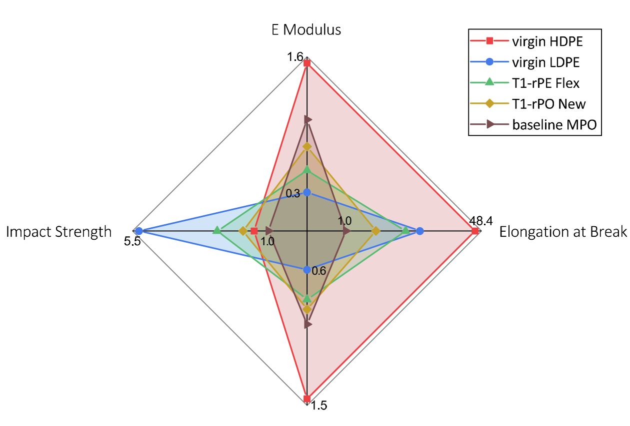

Fig. 5. Mechanical performance comparison between T1-rPE Flex and T1-rPO New regranulates and baseline and reference materials on injection molded specimens.

For the rPE Flex, as example, a successful demonstration is conducted at the company Pezy (The Netherlands) for a connector part, which is welded onto a sturdy PE water bag. Pictures are included in the SI (Fig. SI4). For the rPO New, an example trial is conducted for the injection molding of rooftiles for green roofs. The part has a 3 mm wall thickness and complex geometry. It was previously described in detail by Ragaert et al. (2020). Also this product could be manufactured to satisfaction, of which a picture is included in SI (Fig. SI4).

4. Economic assessment

4.1. Cost-benefit analysis

The detailed comparisons of needed capital investment (CAPEX) and total costs (OPEX), as well as information on the total costs of different parameters (e.g., energy, residual treatment, etc.) can be found in the SI (Figs. SI5- SI7). The CAPEX needed for the process is increased from €24 million in the conventional recycling to €49 and €42 million in scenario 1 and scenario 2, respectively. The annual OPEX for QRP increases from €15 million in the conventional recycling to €26 and €23 million in scenario 1 and scenario 2, respectively. More information and analysis on the economic parameters that drive the increase in CAPEX and OPEX in scenario 1 and scenario 2 can be found in the SI.

Despite the increase in the annual OPEX, it can be observed that QRP improves the net balance of DSD 310–1 and DSD 323–2 recycling (Fig. 6). The negative values indicates the net loss in all scenarios, which gives an important insight into the waste management operation in the market, which to date is not self-sustaining. Yet, our analysis deliberately excludes gate fees, which should be included to assess the final viability of the plants. In fact, many studies suggest that most of the annual OPEX and annualized investment (CAPEX) should be supported by an external source of income (Cimpan et al., 2016; Da Cruz et al., 2014, 2012; Marques et al., 2014). Nevertheless, looking at QRP as an improved mechanical recycling process for DSD 310–1 and DSD 323–2, it can be observed that QRP scenario 2 improves the economic value by 38 % (presented in Fig. 6A). Per bale, the implementation of QRP scenario 2 improves the economic value of processing DSD 310–1 and DSD 323–2 by 57 % and 30 %, respectively. The net loss of processing DSD 310–1 decreases from -€83 per tonne in the baseline to -€36 per tonne in QRP scenario 2, reducing the margin that need to be filled by external parties such as PRO, e.g., via gate fees (Fig. 6B). Similarly, the net loss of processing DSD 323–2 decreases from -€200 per tonne in the baseline to -€141 per tonne in QRP scenario 2 (Fig. 6C). Moreover, recycling of DSD 323–2 shows higher net loss, partly because the bale has a higher contamination level and thus result in a relatively lower yield and generates more residue (Lase et al., 2022). Nonetheless, the economic value of DSD 323–2 recycling is still improved compared to the conventional recycling process. Therefore, these findings indicate that QRP can potentially reduce the external financial support (e.g., gate fees), which is still subjected to further discussion in the circular economy of plastics waste recycling because the financing schemes still vary currently.

4.2. Sensitivity analysis

Many of the cost modelling components can greatly fluctuate. Larrain et al. (2021) have shown that the price of regranulates is amongst others influenced by the oil price. Other components such as energy use or labor cost also vary with time and region (Larrain et al., 2021; PwC, 2019). Therefore, the importance of the selected economic components towards the net profit/loss is investigated through a sensitivity analysis (Fig. 7).

From the results of sensitivity analysis, it can be seen that the price of rPE Film Natural, rPP Film, and rPE Flex are among the most influential parameters, followed by the investment on the selected recycling equipment. This finding indicates the importance of maintaining high

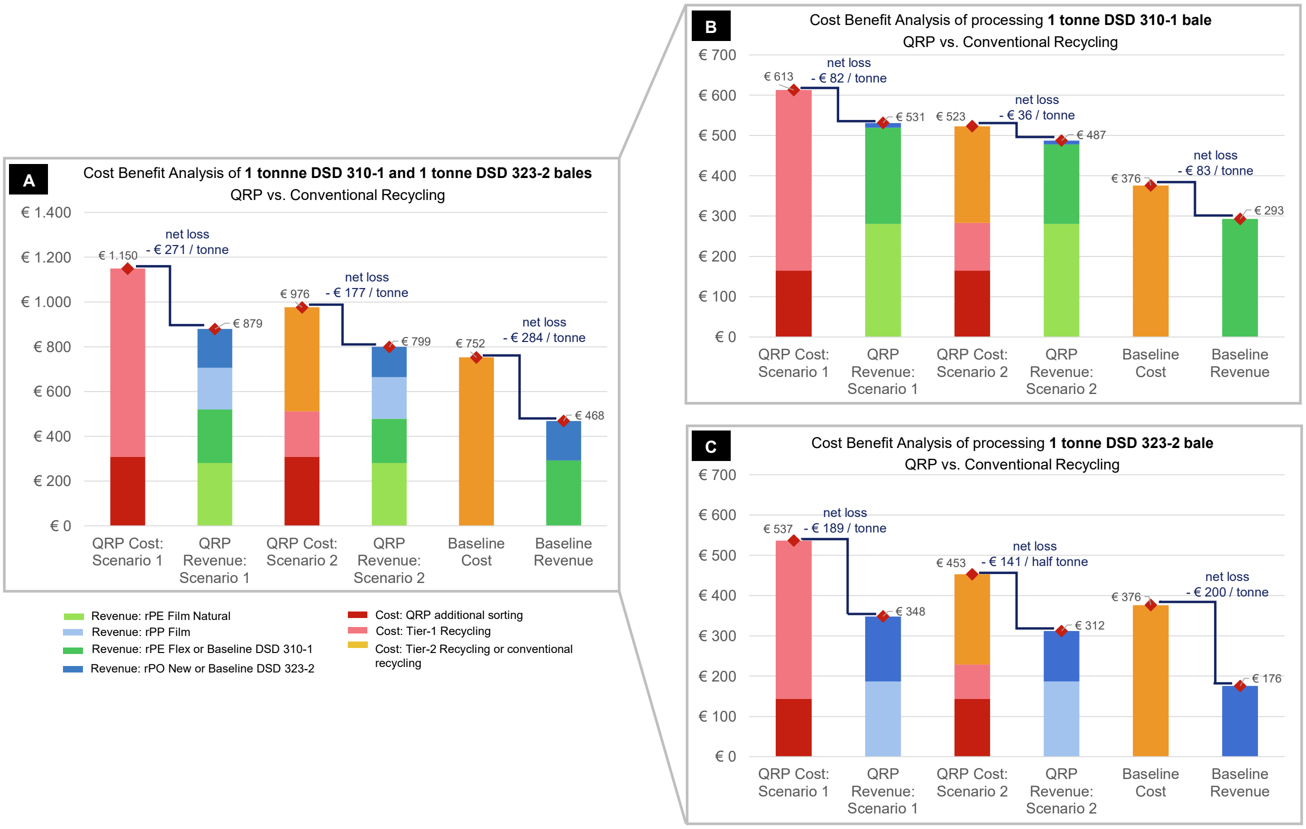

Fig. 6. Net profit/loss of the QRP scenario 1, QRP scenario 2, and baseline scenario: (A) processing 1 tonne DSD 310–1 and 1 tonne DSD 323–2 bales, (B) processing 1 tonne DSD 310–1 bale, and (C) processing 1 tonne DSD 323–2 bale.

quality regranulates, suitable for demanding applications. This also means that a good quality of DSD 310–1 and DSD 323–2 input bales is required so that generated residue can be minimized, thus reducing the cost of residual treatment.

Next, electricity price is the most sensitive parameter followed by depreciation rate. In fact, depreciation accounts for almost one-third of the total cost and ranks amongst the most sensitive parameters. These findings highlight that the strategy on depreciating the investment for each equipment should be properly formulated. The annual OPEX can be significantly reduced if we invest on an equipment with longer lifespan (e.g. equipment that last for 8–10 years). By annualizing the investment to 8–10 years we can see that the economic value can be improved by 18–20 %. Moreover, as the energy and labor costs may differ from one region or country to another (PwC, 2019), it is imperative to make a detailed and regional feasibility study prior to the implementation of QRP.

5. Conclusions

This study evaluates both the quality and economic feasibility of an improved mechanical recycling scheme (QRP) for the recovery of PCFPs. The process innovative improvements are (i) additional sorting units, (ii) a hot wash with a detergent, (iii) improved extrusion with two step filtration, and (iv) deodorization.

The quality of the obtained regranulates is evaluated in this research for film and injection molded applications. It is shown that PE-rich (over 90 % PE) regranulates, on blown films, observe more flexibility (45–60 %), higher ductility (27–55 %), and enhanced tensile strength (5–51 %), compared to the baseline recycling. The dart drop resistance of the recovered regranulates remains typically low and is subject to further investigation. Meanwhile the less pure regranulates exhibit more flexibility (19–49 %), enhanced ductility (7 to 20 times), and higher impact strength (1.8 to 3.8 times), compared to the baseline, indicating improved applicability for injection molding applications. However, due to the lower PP content, the tensile strength decreases (10–16 %). The observed qualities have improved towards the quality of virgin grades, suitable for the same application category. Moreover, PP-rich regranulate, can be extruded into films (unlike the baseline) opening new application possibilities. This was validated in industrial trials, in which the regranulates were blended (at industrially relevant amounts for recyclates) into virgin materials.

Through an economic assessment, it is concluded that the investment and operational cost for the improved process is compensated by delivering more high quality recycled regranulates, which can be used in more diverse applications. It is shown that QRP can be designed in a way that can improve the economic value of the operation by 5–38 %, compared to the conventional recycling. Overall, our results show that it is possible to increase the mechanical recycling quality of PCFP waste in an economically viable way, yet, as in conventional recycling, PRO systems still need to sustain the QRP.

In the opinion of the authors, the main limitations of the current study are: (i) The research was done on a single batch and not duplicated in time, meaning that geographical or seasonal variations in waste composition are not accounted for. (ii) Did not include blending studies to optimize formulations of the regranulated with virgin plastics (iii) Lied on key assumptions of the capital investment (especially in terms of building) and averaged values for some cost modeling parameters (like energy and labor costs) (iv) Did not investigate the effects of further purification steps like finer melt filtration or de-inking. Furthermore, it is strongly advised to read this paper combined with the publication from Lase et al. (2022), who describes the material flow analysis and recycling performance of the same QRP process for PCFP recycling.

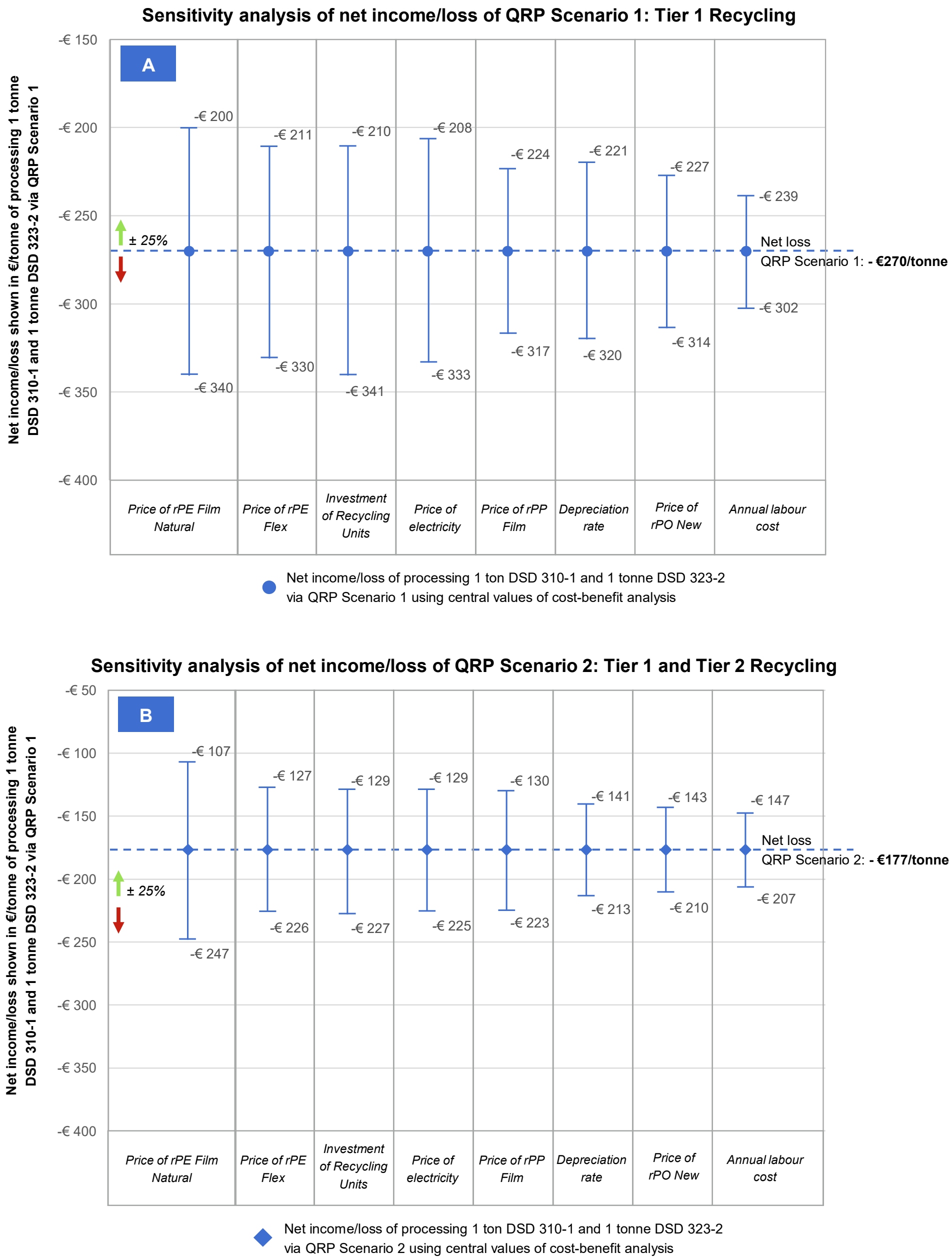

Fig. 7. Sensitivity analysis of the selected cost modeling parameters towards net profit/loss of QRP. In QRP scenario 1 (A), all bales are processed through Tier 1. In QRP scenario 2 (B), PE Film Natural and PP Film bales are processed through Tier 1, whilst PE Flex and PO New bales are processed through Tier 2.

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

No data was used for the research described in the article.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2022.08.018.

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