



Closing the loop for PET, PE and PP waste from households: Influence of material properties and product design for plastic recycling

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ABSTRACT

Recycling of plastic is an important step towards circular economy. However, plastic from household waste (HHW) is a heterogeneous and contaminated resource, leading to recycled plastic with reduced quality, limiting the potential for closed-loop recycling. In addition to regulatory requirements for the chemical composition of recycled plastic, reduced physical and mechanical properties may limit the potential for closed-loop recycling. Consequently, this study analyses the thermal degradation, processability and mechanical properties of a range of reprocessed PET, PE and PP samples from source-separated plastic in HHW. On this basis, the potential for closed-loop recycling is evaluated. The study demonstrated that PET, PE and PP recycling represent different challenges. Potential degradation of the PET polymer can be reversed in a decontamination process, making PET waste well-suited for closed-loop, multiple times recycling, even when the degree of heterogeneity in the waste is high. The processability of different kinds of PE and PP packaging types varied considerably, especially for PP. Consequently, current recycling of mixed PP waste and even separate recycling of individual PP waste packaging types, will not technically facilitate recycling into new packaging products. This highlights the importance of PE and PP waste homogeneity when sent to reprocessing. Such homogeneity may be achieved through additional plastic sorting and regulatory harmonisation of product design, accounting for polymer properties and recyclability. Degradation of PP during recycling was shown to be substantial, representing another important limitation for PP recycling, necessary to address in the future.

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1. Introduction

Plastic materials are essential in modern societies. They are produced mainly from fossil-based resources and in 2016, 335 million tonnes were produced globally (PlasticsEurope and EPRO, 2017), with this figure expected to rise to 1124 million tonnes in 2050 (EMF, 2016). Consequently, recycling of plastic is highlighted as an important step in the transition towards a circular economy (EC, 2015a; EC, 2018), in order to avoid using fossil resources and close the plastic loop. As a result, mass-based recycling targets for packaging plastic and municipal solid waste, including plastic, have been enforced in Europe (EC, 2015b; EU, 2018). However, plastic, especially from household waste (HHW), is very heterogeneous, as it contains many different polymers, product types, product designs, etc. (Eriksen and Astrup, 2019), and it is often contaminated (Ragaert et al., 2017). Consequently, the quality of

the recycled plastic is typically reduced compared to that of the alternative virgin material (Rigamonti et al., 2018; Eriksen et al., 2018a). As the “circularity”, i.e. ability of recycling systems to close material loops, depends strongly on the quality of the recycled material (Eriksen et al., 2018a), potential quality reductions represent a limitation in the transition towards a circular plastic economy (Eriksen et al., 2018a). Thus, to increase the “circularity” of plastic recycling systems, the quality of recycled plastic needs to be improved, preferably to a level allowing closed-loop plastic recycling, i.e. recycling where the plastic material properties are maintained and recycling into similar products is possible (Hopewell et al., 2009).

A major limitation in regards to closed-loop recycling of plastic from HHW is potential presence of unwanted chemicals in the plastic. As the content of additives in different waste products with different purposes can vary greatly, the chemical content of the recycled plastic from mixed plastic waste is very often unknown. Thus, regulatory demands are imposed on the composition of the plastic waste used to produce recycled plastic for food contact

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purposes. As a result the plastic waste input to the final recycling process must contain a minimum of 95% food packaging, in the case of PET, and 100% food packaging, in the case of PE and PP, in order for the waste to be recycled into new food packaging (EC, 2008; EFSA, 2011). As food packaging plastic from HHW is often collected and recycled together with other plastic waste, the recycled plastic is not suitable for conversion into new food packaging, which significantly limits the possibilities for closed-loop recycling. Separate collection and management of food packaging could theoretically solve this and facilitate closed-loop recycling into new food packaging (Eriksen and Astrup, 2019); an illustrative example is separate collection of PET beverage bottles through deposit systems with subsequent recycling into new beverage bottles (Welle, 2011).

However, in addition to chemical and legal restrictions, mechanical and physical properties may further limit the potential for closed-loop recycling. First, because important properties might be reduced during recycling. As an example, several studies have reported that the mechanical properties of recycled plastic, especially when originating from HHW, were reduced when compared to virgin plastic. This includes tensile strength and elongation to break for recycled PP (Dahlbo et al., 2018), tensile strength of recycled HDPE (Luijsterburg and Goossens, 2014; Chariyachotilert et al., 2006), elongation to break for recycled LLDPE (Al-Salem, 2019), and impact strength of recycled PP (Barbosa et al., 2017). These changes in properties might be caused by thermal-mechanical degradation of the polymer during reprocessing, degradation during the lifetime, mainly due to photo oxidation processes (Ragaert et al., 2017), and/or contamination of the plastic waste, e.g. in cases with complex product designs involving several immiscible polymers or materials (Hahladakis and Iacovidou, 2018). Second, because the same polymer used in different product types (bottles, trays, tubs, etc.) can exhibit different properties. For instance, the processability of different product types may vary significantly. The processability is an important property for the industry, as it determines which production method is applicable for PE and PP. As an example, plastic with low processability is suitable for blow moulding into bottles, whereas high processability is suitable for injection moulding into tubs or buckets with thin walls (Frederiksen, 2018; Scholdan, 2018). The processability of recycled plastic from mixed waste, containing many different packaging types originally exhibiting different processability, might therefore limit the potential for closed-loop recycling into similar packaging products (bottles, trays, etc.). While closed-loop recycling might be possible through dilution with virgin plastic (Ragaert et al., 2017) and open-loop recycling into other product types with other requirements is certainly possible, such situations represents physical losses of material or loss of material properties, both of which lowers the overall “circularity” of the plastic system. These types of recycling are therefore not considered in this study.

To improve the “circularity” of plastic recycling, separation between individual polymer types is certainly required (Ragaert et al., 2017). For HHW this most often includes separation of the three most dominant polymers, PET, PE and PP (Feil et al., 2017; Eriksen and Astrup, 2019). Moreover, separation between food and non-food packaging is important from both a chemical and regulatory perspective. Finally, separation between individual product types might be critical for the processability and mechanical properties of the recycled plastic. Several studies have previously demonstrated the importance of effective polymer separation for recovering large quantities of plastic waste with low levels of physical contamination (e.g. Faraca et al., 2019; Eriksen et al., 2018a). Moreover, Eriksen and Astrup (2019) have demonstrated that separate recycling of food-packaging waste is crucial in order to facilitate closed-loop recycling of food-packaging from a chemical and

a regulatory perspective. However, even with effective polymer separation, where food packaging is recycled separately, it has not previously been assessed how the distribution of product types in plastic waste may influence the physical and mechanical properties of recycled plastic – and thereby how to facilitate closed-loop recycling of plastic from HHW from a physical and mechanical perspective.

Eriksen and Astrup (2019) provided a detailed composition of Danish, source-separated, rigid plastic waste. They found that plastic beverage bottles, trays for fruit and vegetables, meat trays, dairy tubs, additional mixed trays for food-purposes and bottles for soap-related purposes (i.e. containers for detergents, shampoo, etc.) all represented a considerable share of the waste. As these packaging types may exhibit different material properties, they are highly relevant, when understanding how the composition of plastic from HHW may influence the material properties of recycled plastic. Moreover, the study revealed that several of these waste packaging types consisted of multiple polymers. If these polymers are not sufficiently separated during recycling via e.g. density separation, representing the most dominant method for separation of plastic flakes (Ragaert et al., 2017), such multi-polymer product designs might additionally limit the quality of the reprocessed plastic (Hahladakis and Iacovidou, 2018). Reductions in quality that could be avoided by changing to mono-polymer product designs, as suggested by several organisations (Rethink Plastic, 2018; FCP, 2018; APR, 2018). Furthermore, current recycling practises involve reprocessing of mixed PET, PE and PP waste (Eriksen and Astrup, 2019) and thus, the material properties of reprocessed plastic from mixed waste is crucial, in order to evaluate the current situation. Thus, systematic assessment of the relationship between plastic waste composition, focussing on these packaging types, and material properties of recycled plastic is necessary in order to evaluate if, and to what extent, separate management of specific packaging types, specific sorting processes and product design changes can improve the basis for closed-loop plastic recycling, ultimately increasing the overall “circularity” of the plastic system.

The aim of the study was to analyse a range of reprocessed plastic samples, representing different potential waste management options and product designs, with respect to selected physical and mechanical properties, and on this basis assess the influence of these properties on the potential for closed-loop recycling of plastic from household waste. This involved the following specific objectives: (1) collection of source-separated PET, PE and PP waste from households, (2) preparation of reprocessed plastic samples, by shredding, washing, extrusion and pelletising, (3) analyse the thermo-gravimetric properties, melt flow index (MFI), tensile properties and impact strength of each sample, (4) evaluate the potential of mechanical closed-loop recycling of each sample, and on this basis (5) provide recommendations on how to improve the “circularity” of plastic recycling from household waste.

2. Methodology

2.1. Samples

This study included 25 reprocessed PET, PE and PP samples, in the form of pellets, representing raw plastic material after source-separation, collection, sorting and reprocessing but before conversion into products. Nineteen of the samples originated from Danish source-separated plastic waste collected from households in Copenhagen in 2017 and were prepared following the procedure described in Section 2.2. The remaining six samples were collected from industrial scale recycling companies from Denmark, Germany and the Netherlands (sample name ends with .ExX), and have

previously been analysed for the metal concentration in Eriksen et al. (2018b). The external samples were collected in the form of pellets and were therefore analysed as received.

An overview of all samples, including the names used in the remaining paper, is presented in Table 1. Additionally, Table 1 provides an overview of what the samples represent: (1) Mixture of food and non-food Products (MP), (2) samples made from Specific waste Packaging types (SP), (3) waste packaging types manually modified to represent a specific change in the Product Design (PD) or (4) samples that have been through an active Density Separation process (DS). Samples of mixed plastic waste (including food and non-food packaging and non-packaging) were included, to represent current recycling conditions. Samples of individual waste packaging types were included to quantify potential differences in material properties between different packaging types, and on that basis evaluate if separate collection or recycling could facilitate closed-loop recycling. Moreover, individual waste packaging types, known from Eriksen and Astrup (2019) to consist of multiple polymers, were manually manipulated to simulate a

mono-polymer design. These samples were included to quantitatively evaluate how potential design changes to mono-polymer products might influence the material properties of the recycled plastic. Finally, samples partitioning into a floating and a sinking fraction during washing were subjected to a density separation process, in order to evaluate the influence of such processes.

2.2. Sample preparation

The waste used to prepare the PET, PE and PP samples of individual packaging types were collected as part of the waste characterisation campaign, described in details in Eriksen and Astrup (2019). In this campaign rigid, source-separated plastic waste from the municipality of Copenhagen, was mechanically sorted into PET, PE and PP waste. Each of these fractions were manually mixed before a primary sample of 5–10 kg was obtained. Beverage bottles, trays for fruit and vegetables and additional mixed food trays were manually separated from the primary PET sample, bottles for soap related purposes was manually separated from the primary

Table 1

Sample overview with details related to origin and treatment. The sample names will be used in the remaining paper. HHW: Household waste, MP: Mixed Products, SP: Specific waste Packaging types, PD: Product Design, DS: Density Separation, PET: Polyethylene terephthalate, PE: Polyethylene, PP: Polypropylene.

| Sample | Polymer | Waste packaging type | Waste origin and collection | Additional pretreatment ¹ | | Sample representing | | | |
|--------------------|---------|---------------------------|-----------------------------|--------------------------------------|--------------------------|---------------------|----|----|----|
| | | | | Manual removal of | Density separation | MP | SP | PD | DS |
| PET.MixFoodTray | PET | Mixed food trays | Danish source-separated HHW | – | – | ✓ | | | |
| PET.Fruit.DS | PET | Fruit and vegetable trays | Danish source-separated HHW | – | ✓ Removal mainly of pulp | ✓ | | | ✓ |
| PET.Fruit.All | PET | Fruit and vegetable trays | Danish source-separated HHW | – | – | ✓ | | | |
| PET.BevBottle.NoLL | PET | Beverage bottles | Danish source-separated HHW | ✓ Plastic lids and labels | – | ✓ | ✓ | | |
| PET.BevBottle.DS | PET | Beverage bottles | Danish source-separated HHW | – | ✓ | ✓ | | | ✓ |
| PET.BevBottle.All | PET | Beverage bottles | Danish source-separated HHW | – | – | ✓ | | | |
| PET.Mix.DS | PET | Mixed products | Danish source-separated HHW | – | ✓ | ✓ | | | ✓ |
| PET.Mix.All | PET | Mixed products | Danish source-separated HHW | – | – | ✓ | | | |
| PE.SoapBottle.NoLL | PE | Bottles for soap purposes | Danish source-separated HHW | ✓ Plastic lids and labels | – | | ✓ | ✓ | |
| PE.SoapBottle.All | PE | Bottles for soap purposes | Danish source-separated HHW | – | – | | ✓ | | |
| PE.Mix | PE | Mixed products | Danish source-separated HHW | – | – | ✓ | | | |
| PE.Ex1 | PE | Mixed products | Unknown HHW | Unknown ² | Unknown ² | ✓ | | | |
| PE.Ex2 | PE | Mixed products | Unknown HHW | Unknown ² | Unknown ² | ✓ | | | |
| PE.Ex3 | PE | Mixed products | Unknown HHW | Unknown ² | Unknown ² | ✓ | | | |
| PP.Dairy | PP | Dairy tubs | Danish source-separated HHW | – | – | | ✓ | | |
| PP.MixFoodTray | PP | Mixed food trays | Danish source-separated HHW | – | – | | ✓ | | |
| PP.Fruit | PP | Fruit and vegetable trays | Danish source-separated HHW | – | – | | ✓ | | |
| PP.Meat.NoFoil | PP | Meat trays, raw meat | Danish source-separated HHW | ✓ Plastic sealing foil | – | | ✓ | ✓ | |
| PP.Meat.DS | PP | Meat trays, raw meat | Danish source-separated HHW | – | ✓ Removal mainly of pulp | | ✓ | | ✓ |
| PP.Meat.All | PP | Meat trays, raw meat | Danish source-separated HHW | – | – | | ✓ | | |
| PP.Mix.DS | PP | Mixed products | Danish source-separated HHW | – | ✓ | ✓ | | | ✓ |
| PP.Mix.All | PP | Mixed products | Danish source-separated HHW | – | – | ✓ | | | |
| PP.Ex1 | PP | Mixed products | Unknown HHW | Unknown ² | Unknown ² | ✓ | | | |
| PP.Ex2 | PP | Mixed products | Unknown HHW | Unknown ² | Unknown ² | ✓ | | | |
| PP.Ex3 | PP | Mixed products | Unknown HHW | Unknown ² | Unknown ² | ✓ | | | |

¹ Besides shredding, washing, drying, extrusion and pelletisation.

² The entire sorting and recycling process is unknown.

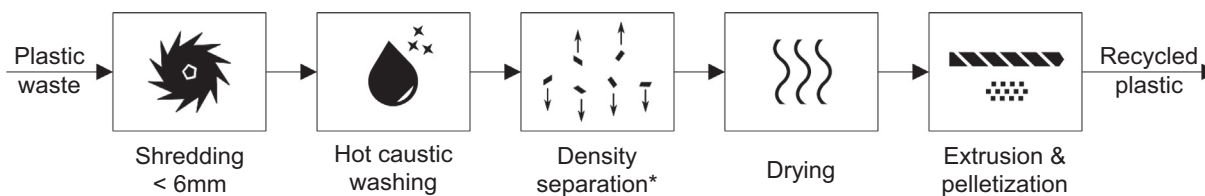


Fig. 1. Sample preparation procedure. * was only performed on selected samples, see Table 1.

PE sample and dairy tubs, trays for fruit and vegetables, meat trays and additional mixed food trays were manually separated from the primary PP sample. The samples of PET beverage bottles, PE bottles for soap related purposes and PP meat trays were further divided in two, where one stayed unchanged and one was manipulated through removal of either plastic lids and labels or sealing foil, to simulate a mono-polymer product design.

The waste used to prepare the samples of mixed PET, PE and PP was similarly obtained from rigid, source-separated plastic waste from the municipality of Copenhagen, after mechanical sorting into PET, PE and PP. 10 kg of each polymer fraction was collected after manual mixing, coarsely shredded and then mixed again, before around 1 kg was collected for final preparation.

This resulted in 14 samples of 0.5–1.5 kg, all of which were subsequently prepared following the procedure illustrated in Fig. 1 and described in detail in the following sections. Visual presentation of the samples before, during and after preparation is provided in Appendix A2 in the supplementary material (SM).

2.2.1. Shredding

All samples were shredded into flakes <6 mm during one cycle, using a flat plastic crusher with rotating blades provided by Dongguan Naser Machinery Co., Ltd, model NPCP-50.

2.2.2. Washing, density separation and drying

To simulate a generic washing process at industrial scale reprocessing facilities, all samples were washed in highly agitated, hot and caustic conditions for 15 min.

After washing, the plastic flakes were transferred to a large, transparent beaker, and in cases where the material separated into a sinking and a floating fraction, the sample was divided in two; one containing both the sinking and floating fraction (sample name ends with .All) and one containing only the targeted fraction of the two (sample name ends with .DS). The specific washing and density separation procedure is presented in Appendix A1 in the SM and was based on several guidance documents from the Association of Plastic Recyclers (APR, 2018). Finally, all samples were dried at 60 °C for 24 h.

2.2.3. Extrusion and pelletisation

Plastic flakes were automatically fed into a PRISM Eurolab 16 twin-screw counter rotation extrusion machine with six temperature-controlled zones and a screw configuration with a decompression zone in the middle. The extruded plastic string was cooled in a stainless steel water bath at room temperature and cut into pellets by an L-002-1345 pellet cutter with a rotation knife.

The extrusion conditions were equal for all samples of the same polymer, see Table 2. The conditions under which the samples were extruded were defined by experimenting with temperature and frequency of rotation, starting from the minimum processing temperature reported for the specific polymer, and increasing until the plastic flowed unhindered.

Table 2

Extrusion conditions in all six temperature-controlled zones.

| Samples | Temperature [C] | Frequency of rotation [rpm] |
|-------------|-----------------|-----------------------------|
| PET samples | 265 | 100 |
| PE samples | 230 | 200 |
| PP samples | 200 | 50 |

2.3. Material property testing

2.3.1. Thermo-gravimetric analysis (TGA)s

Analysis of the degradation pathway was performed on a Discovery TGA instrument from TA Instruments, on samples of 10–20 mg in accordance to instrument recommendations. Each sample was tested in a nitrogen atmosphere, ranging from room temperature up to 700C with a heating rate of 10 K/min. From this analysis, the following properties were determined:

- Temperature [C] at 5% and 10% degradation ($T_{D,5\%}$, $T_{D,10\%}$)
- Slope of degradation [%/C], assuming a linear degradation between 30 and 70, i.e. one weight loss step
- Residues left after 700C [%]

In a very uniform sample, the degradation temperatures will be close to each other and the slope will be low, as this indicates fast degradation. On the other hand, early degradation and a higher slope suggests slower degradation over a wider temperature range, indicating the presence of more compounds.

2.3.2. Melt mass-flow index (MFI)

The melt mass-flow index (MFI) is a measure of the processability of plastic. It is a crucial property for especially PE and PP plastic, as the plastic's suitability for different processing methods are strongly influenced by the MFI. Consequently, the MFI of all PE and PP samples was analysed according to ISO 1133, method A (cutting off and weighing method), under the conditions presented in Table 3, using an MFI 10 Melt Flow Indexer from METTEK.

2.3.3. Tensile test

Tensile testing was performed on an Instron 5542 machine with a 2-kN load cell. All testing was performed on ISO 3167 test specimens of $9.9 \times 3.75 \text{ mm}^2$, approved for the ISO 527-2 tensile testing procedure, with a tensile rate of 50 mm/min and gauge separation of 26.0–28.1 mm. The tensile strength and strain were defined at break for PET samples and at yield for PE and PP samples. Five test specimens of each sample were tested, and the tensile strength and strain were determined from an average between these.

Table 3

Melt mass-flow index (MFI) analysis conditions.

| Polymer | Temperature [C] | Load [kg] | ISO-standard |
|---------|-----------------|-----------|---------------------|
| PE | 190 | 5 | 1872-1, condition T |
| PP | 230 | 2.16 | 1873-1, condition M |

2.3.4. Impact test

Unnotched Izod impact testing was performed on an Instron CEAST 9050 machine with a DAS 8000 Junior data-acquisition system, a sample frequency of 1000 kHz and a 50-J instrumented pendulum hammer. The tests were performed on specimens 80x10x4 mm in size, following ISO180. The impact strength was found as average values between five specimens for PP and PE samples, and three for PET samples.

3. Results and discussion

3.1. Thermo-gravimetric analysis

The degradation properties for all samples are presented in Table 4. All degradation curves, with the exception of those presented in Fig. 2, are presented in Appendix B1 in the SM.

3.1.1. PET

Between 12.1 and 15.3 % residue was left in all PET samples after heating to 700 °C, representing a considerable amount. This is mainly ascribed to ash formation due to incomplete degradation of the aromatics in PET, which is a well-known phenomenon, in line with levels generally observed for virgin unfilled PET (Adnan et al., 2015).

The degradation profiles of all PET samples were very similar, and only small variations between samples were observed. The two samples containing other polymers from lids and labels (PET.BevBottles.All and PET.Mix.All) had a slope of $-1.7\%/^{\circ}\text{C}$, which was $0.1\text{--}0.2\%/^{\circ}\text{C}$ higher than any of the other samples, potentially indicating that they were less chemically uniform, as would be expected. However, the results clearly indicate that potential polymer cross-contamination has little to no influence on the degradation profile of these PET samples.

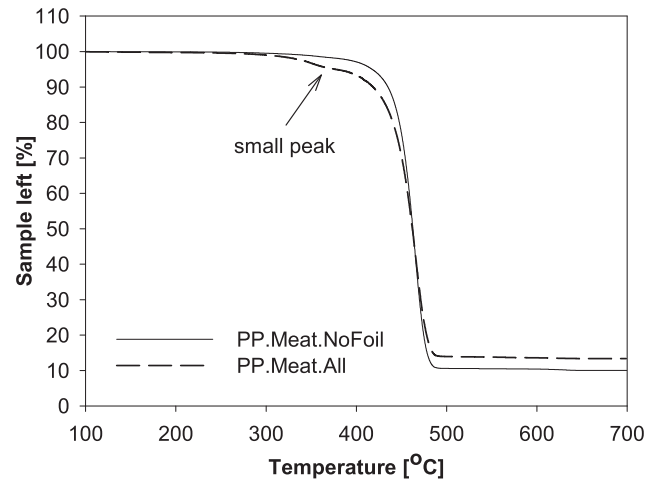


Fig. 2. Thermo-gravimetric degradation of reprocessed PP from waste meat trays with (PP.Meat.All) and without (PP.Meat.NoFoil) sealing foil.

3.1.2. PE

Some differences were observed for the PE samples, as 5–10% degradation happened at slightly higher temperatures for bottles without labels and lids (PE.SoapBottle.NoLL), when compared to samples including labels and lids (PE.SoapBottle.All and PE.MIX). This indicates that the sample representing a mono-polymer design was the most homogeneous, and that lids and labels made from other polymers, or a different grade of PE, contribute to the chemical heterogeneity of reprocessed PE.

The highest $T_{D,5\%}$ and $T_{D,10\%}$, and the lowest degradation slopes, were observed for two of the external samples (PE.Ex2 and PE.Ex3), demonstrating the fastest and most uniform degradation of all PE

Table 4

Degradation properties of all samples obtained from thermo-gravimetric analysis. $T_{D,5\%}$: Temperature at 5% degradation, $T_{D,10\%}$: Temperature at 10% degradation.

| Samples | $T_{D,5\%}$ [°C] | $T_{D,10\%}$ [°C] | Residue ¹ [%] | Slope ² [%/°C] |
|---|---------------------|----------------------|-----------------------------|------------------------------|
| Polyethylene terephthalate (PET) | | | | |
| PET.MixFoodTray | 399 | 410 | 15.3 | −1.8 |
| PET.Fruit.DS | 398 | 410 | 13.4 | −1.9 |
| PET.Fruit.All | 398 | 410 | 13.2 | −1.9 |
| PET.BevBottle.NoLL | 400 | 410 | 14.1 | −1.8 |
| PET.BevBottle.DS | 400 | 410 | 13.9 | −1.9 |
| PET.BevBottle.All | 401 | 411 | 12.8 | −1.7 |
| PET.Mix.DS | 399 | 409 | 12.1 | −1.9 |
| PET.Mix.All | 397 | 408 | 13.7 | −1.7 |
| Polyethylene (PE) | | | | |
| PE.SoapBottle.NoLL | 444 | 457 | 1.7 | −3.0 |
| PE.SoapBottle.All | 438 | 451 | 1.4 | −2.7 |
| PE.Mix | 439 | 453 | 1.8 | −2.9 |
| PE.Ex1 | 438 | 450 | 0.9 | −2.6 |
| PE.Ex2 | 464 | 470 | 1.2 | −3.7 |
| PE.Ex3 | 465 | 471 | 0.8 | −3.9 |
| Polypropylene (PP) | | | | |
| DairyPP.Dairy | 406 | 425 | 2.4 | −2.4 |
| PP.MixFoodTray | 413 | 428 | 1.4 | −2.5 |
| PP.Fruit | 406 | 423 | 0.6 | −2.3 |
| PP.Meat.NoFoil | 418 | 435 | 10.0 | −2.5 |
| PP.Meat.DS | 398 | 424 | 13.3 | −2.1 |
| PP.Meat.All | 377 | 419 | 13.4 | −1.9 |
| PP.Mix.DS | 406 | 424 | 2.4 | −2.4 |
| PP.Mix.All | 408 | 426 | 2.1 | −2.4 |
| PP.Ex1 | 425 | 438 | 1.6 | −2.8 |
| PP.Ex2 | 415 | 431 | 1.9 | −2.6 |
| PP.Ex3 | 420 | 433 | 2.1 | −2.6 |

¹ Left after 700 °C

² Calculated between 30 and 70% degradation.

samples, thereby representing the most chemically homogeneous samples. However, as no information was available on either the recovery or reprocessing, it was not possible to pinpoint exactly to what this may relate.

3.1.3. PP

The samples consisting of meat trays degraded differently from the rest of the PP samples, as the share of inorganic residues left after 700°C was considerably higher (10–13.4%) when compared to the remaining samples (0.6–2.4%). This suggests that various additives, possibly inorganic fillers, may have been used in the production of meat trays, representing a substantial share of the mass. Moreover, differences were observed when comparing the three meat tray samples. Samples including sealing foils (*PP.Meat.DS* and *PP.Meat.All*) exhibited very similar levels of degradation and contained higher residues, 13.3% and 13.4%, compared to 10% for the sample without sealing foils (*PP.Meat.NoFoil*), see Fig. 2. [INCEN \(2011\)](#) reported that the main part of sealing foils for meat trays is often made of PET, which could explain the increased amount of inorganic residues in the samples including sealing foils, as ash from aromatic compounds in the PET is expected to remain as inorganic residues – as demonstrated for the PET samples. Moreover, the samples including sealing foils exhibited a small peak in degradation at around 350°C, as indicated in Fig. 2. Sealing foils are often coated with EVOH or nylon ([INCEN, 2011](#)); this peak may therefore originate from these two materials or indeed PET. In any case, the extra degradation peak indicates that sealing foils consist of material(s) chemically different from the PP in the tray. A chemical heterogeneity unlikely to be reduced by density separation, as *PP.Meat.DS* and *PP.Meat.All* exhibited similar degradation.

3.2. Processability

The MFI, a widely used measure for the processability, determines the appropriate processing method for PE and PP plastic and thus, which product types are suitable for a given PE and PP material. This correlation is presented in Fig. 3 based on experience from the Danish recycling industry ([Scholdan, 2018](#)). The figure shows that plastic with MFI values between 0 and 1 g/10 min is suitable for extrusion, which is often used to produce plastic bags or film, thermoformed into low trays. Slightly higher MFI values, between 0.3 and 5 g/10 min, are suitable for blow moulding, used to produce bottles. MFI values from 5 g/10 min and upward are suitable

for injection moulding. MFIs between 5 and 50 g/10 min can be used to mould thicker-walled products, such as toys or outdoor fences, but not most packaging products, as they are often thin-walled products, which requires MFIs above 50 g/10 min. Fig. 3 illustrates how the MFIs of the PE and PP samples were distributed. Specific MFI-values are provided Appendix B3 in the SM.

3.2.1. PE

All PE samples exhibit MFI-values below 9 g/10 min, and the samples specifically prepared for this specific study (*PE.SoapBottle.NoLL*, *PE.SoapBottle.All* and *PE.Mix*) had very similar MFIs, between 0.8 and 1 g/10 min. As soap related bottles were expected to originally be produced exclusively by blow moulding, requiring MFIs between 0.3 and 5 g/10 min, potential degradation of the polymer during recycling, leading to an increase in MFI, does not seem to be pronounced for *PE.SoapBottle.NoLL* and *PE.SoapBottle.All*. This is in line with an earlier study concluding that PE is suitable for multiple extrusions, as the MFI is relatively unchanged when the processing temperature remains below 240 °C ([Kozłowski, 2015](#)). Hence, the MFI of reprocessed PE from soap bottles technically allows closed-loop recycling into new bottles. Even reprocessed PE from mixed PE waste (*PE.Mix*) can be recycled into new bottles. Since different PE products (such as bottles, tubs and dishes) are made from different PE grades, known to differ considerably in MFI ([Kozłowski, 2015](#)), this is a surprising observation as mixed plastic waste often contains many different product types. However, the source-separated PE waste, used to produce the mixed PE sample, consisted of around 80% bottles ([Eriksen and Astrup, 2019](#)), which is expected to be the reason why samples of mixed PE perform very similar to those made exclusively from soap related bottles. Thus, the homogenous nature of the mixed PE waste is an advantage during recycling, highlighting the importance of a homogenous PE waste stream for closed-loop recycling.

The external samples made from PE from HHW varied substantially more in MFI, from 0.3 to 9 g/10 min. These samples were collected from different geographical locations, where the distribution of packaging types within the PE waste might be different from the distribution in the Danish source-separated rigid PE waste. Moreover, the presences of PP in a PE stream might increase the MFI, as reprocessed PP in general exhibited higher MFIs than reprocessed PE (see Fig. 3). The high MFI of *PE.Ex2*, could therefore be due to presence of PE products, made from different PE grades, or PP products, making the reprocessed PE unsuitable for bottle production, further highlighting the importance of a homogeneous PE waste stream.

3.2.2. PP

The MFIs of the PP samples varied considerably, from 3 g/10 min to 79 g/10 min.

Reprocessed PP produced from meat trays had the lowest MFIs of 3–4 g/10 min. This indicated that a large share, if not all, of the PP meat trays were originally produced by extrusion and thermoforming, as this requires MFI between 0 and 1 g/10 min. The slightly higher MFIs for the reprocessed meat tray samples are most likely due to the expected degradation of PP during reprocessing, known to increase the MFI ([Kozłowski, 2015](#); [Delva et al., 2014](#)), thereby making recycling of meat trays unsuitable for closed-loop recycling into new meat trays produced with extrusion and thermoforming. Moreover, the plastic string produced from these samples had a rough surface, not smooth as for the other samples. Following visual inspection, these samples therefore had a low quality, potentially due to the high share of inorganic fillers, demonstrated by the TGA, representing a general limitation for plastic recycling.

As opposed, reprocessed PP from dairy tubs had the highest MFI of all samples of 79 g/10 min, indicating that dairy tubs were orig-

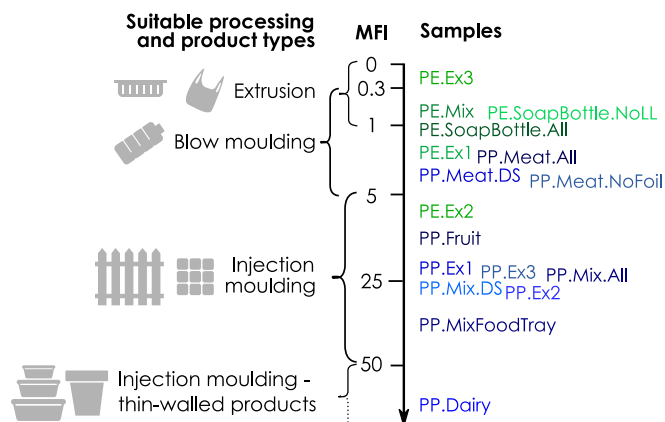


Fig. 3. Melt flow index (MFI) scale, linking MFI of PE and PP plastic to suitable processing methods and typical types of products produced from these methods, as well as illustrating the distribution of MFI-values for the samples in this study. PE samples are indicated in green, PP samples in blue. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

inally produced exclusively from injection moulding suitable for the production of thin-walled products, requiring MFI > 50 g/10 min. Thus, despite expected degradation, separate recycling of dairy tubs seems to produce reprocessed PP technically able to facilitate closed-loop plastic recycling. However, when the polymer chains are cleaved as a result of processing, the PP also becomes more brittle (Kozłowski, 2015), which might be an issue after recycling multiple times.

As expected, all of the reprocessed PP samples from mixed PP waste exhibited MFIs in the same range (21–27 g/10 min), representing an average between extruded trays, e.g. meat trays, and thin-walled injection moulded containers, such as dairy tubs. Consequently, the MFI of reprocessed PP from mixed waste only allowed for injection moulding of thick-walled products, such as toys, traffic cones, outdoor fences, etc. On the contrary, it was not expected that *PP.MixFoodTray* and especially *PP.Fruit* would also exhibit MFIs between 5 and 50 g/10 min, making them unsuited for closed-loop production into new trays and tubs, as these samples were made from more homogenous waste packaging types. Hence, the results indicate that trays and tubs, such as fruit and vegetable packaging, are most likely not produced in the same way although the products share the same purpose, representing a major challenge for closed-loop recycling, even if collected and recycled separately from the remaining waste.

3.3. Mechanical properties

Tensile strength [MPa], tensile strain [%] and impact strength [kJ/m²] are presented for all samples in Fig. 4. All stress-strain curves are presented in Figs. B4–B6 in the SM.

3.3.1. PET

The tensile strength of the reprocessed PET was between 12.9 and 34.6 MPa and were thereby considerably reduced compared to virgin PET, having typical tensile strength between 50 and 80 MPa (Crompton, 2012; PP, 2018). Moreover, the impact strength of the reprocessed PET samples was between 2.2 and 10 kJ/m², representing a dramatic reduction compared to the impact strength of virgin PET, which typically lies between 130 and 160 kJ/m² (MatWeb, 2018). Additionally, Fig. 4 shows that the relative variation in both tensile strength and strain at break was substantial. Thus, the reprocessed PET samples did clearly not represent desirable raw material, and they may possibly not be suitable for any applications.

Awaja and Pavel (2005) reported that the most important factor affecting the suitability of post-consumer PET plastic for recycling is the level of contamination. As such, it was expected that samples where lids and labels of other polymers were manually removed (*PET.BevBottle.NoLL*), or samples from mono-polymer PET products (*PET.MixFoodTray*, *PET.Fuit.DS*), would exhibit higher mechanical properties, compared to samples including polymer cross-contamination from lids and labels (*PET.BevBottles.All* and *PET.Mix.All*). However, the results obtained from the PET samples did not confirm this trend and it was therefore not possible to conclude any systematic influence from contamination on the mechanical properties of these reprocessed PET samples.

In addition to contamination, proper drying of the PET waste before (re)processing has been reported as crucial for the quality of the reprocessed PET (Awaja and Pavel, 2005; Kozłowski, 2015). Where the MFI is crucial for the recyclability of PE and PP plastic, the intrinsic viscosity (IV), a measure of the length of the polymers, is crucial for the applicability of PET plastic, as bottle production requires higher IV than e.g. tray and film production. During (re) processing, hydrolysis reactions will occur between water and PET, breaking the PET polymers and thereby reducing the IV. If the moisture content is above 0.02%, PET will immediately degrade

during processing, often resulting in catastrophic reductions in the IV and as a consequence the mechanical performance (Sepe, 2012). As the initial humidity of PET flakes has been reported at 0.7% (Kozłowski, 2015), and the reprocessed PET samples analysed in this study were not subjected to a dedicated drying process, the moisture content of the PET waste was most likely substantially above 0.02% when entering the extrusion process (Stoughton, 2014). The low mechanical properties and large variations between replicates thus supports the importance of a dedicated drying process during PET recycling and the mechanical properties of the reprocessed PET samples are therefore not expected to represent those of PET reprocessed at industrial scale, as industrial-scale reprocessing facilities have dedicated drying processes before, and in some cases, during extrusion (Lynggaard, 2018; Awaja and Pavel, 2005).

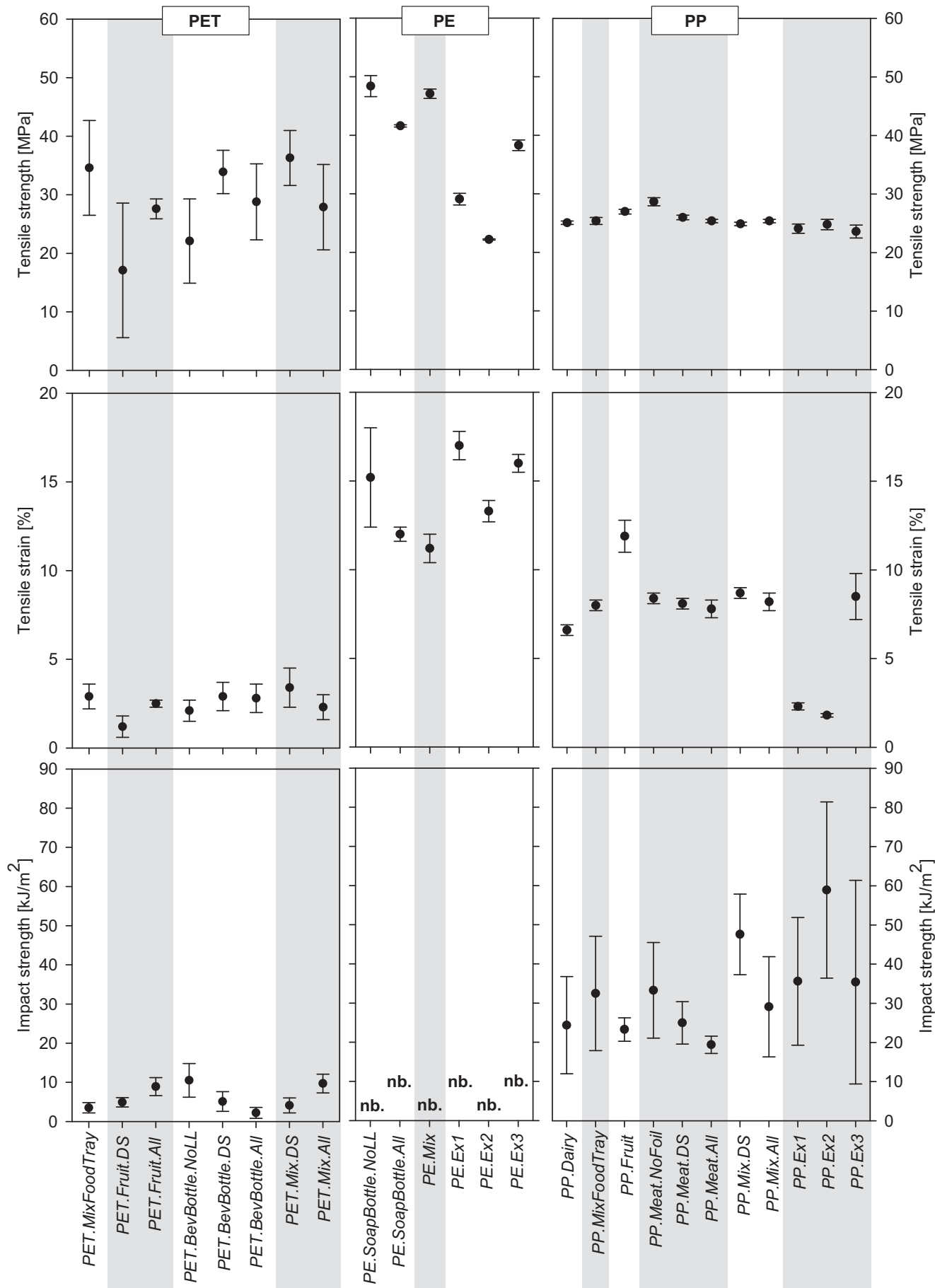
Recycled PET intended for use in food contact applications needs to go through a decontamination process to ensure that potentially unwanted substances are effectively removed to levels not representing a risk to human health (EC, 2008). Hydrolysis of the PET polymer, which degraded the polymers in the first place, is a reversible process (Kozłowski, 2015) and thus, the polymers and the IV of PET plastic can be rebuild during such decontamination processes. As a result, recycled PET can achieve quality criteria sufficient for use in bottles for food contact (Rieckmann et al., 2011), even when the waste consist of PET trays or film, expected to have an IV unsuitable for bottle production before the decontamination process (Kozłowski, 2015). Consequently, the distribution of PET packaging types in the waste might ultimately have little or no influence on the material quality of the reprocessed PET for food contact purposes. The level of degradation in waste products might, however, influence the length and/or temperature of the decontamination process and thereby affect the environmental impacts from a life cycle perspective, an aspect which is beyond the scope of this study.

As such, the results highlight the importance of effective decontamination in order to sufficiently rebuild the polymer chains, as well as effective water removal, limiting the degradation of the polymers during processing and subsequent conversion into products.

3.3.2. PE

The tensile strengths were between 42 and 48 MPa for reprocessed samples prepared for this study, which is extraordinary high, compared to tensile strength for virgin HDPE of 20–32 MPa, found using the same tensile rate as in this study (Koyun et al., 2012). This could be because the processed PE samples were made primarily from bottle-grade HDPE with low MFIs, see 'Section 3.2.1. Low MFI indicate long polymer chains, known to result in high tensile strength. However, as PE is a crystalline polymer, the specific crystallinity of the plastic might also affect mechanical properties. In summary, the tensile properties did not seem to reduce during recycling, which is in accordance with the findings of Kozłowski (2015). Furthermore, contamination from lids and labels had little influenced on the mechanical properties.

As illustrated in Fig. 4, the external reprocessed samples of mixed PE products from HHW (*PE.Ex1*, *PE.Ex2* and *PE.Ex3*) exhibited tensile strength considerable lower, at 22.3–38.6 MPa, lying within the typical range for virgin HDPE. As the composition of the PE waste used to produce the external samples (*PE.Ex1*, *PE.Ex2* and *PE.Ex3*) was unknown, and it is normal practice in many countries to source-separate rigid and flexible plastic together (e.g. van Velzen, 2013), it is reasonable to assume that the reduced tensile strengths could be due to a higher degree of product mixing, most likely from some degree of flexible LDPE, having a typical tensile strength of only 10 MPa (Crompton, 2012). The packaging types



in PE waste do therefore seem to have a considerable influence on the mechanical properties of reprocessed PE.

3.3.3. PP

As illustrated in Fig. 4, the tensile strength of all the reprocessed PP samples lied within a narrow range of 24–29 MPa, which is at the lower end of what is typical for virgin PP, i.e. 21–40 MPa (Klein, 2011; PP, 2018). Moreover, Klitkou et al. (2011) reported an impact strength of 47 kJ/m² for virgin PP, and as the impact strength of the reprocessed samples varied from 19 to 47 kJ/m², with a single external samples of 59 kJ/m², the impact strengths observed in this study were also at the lower end. This could be due to the inevitable degradation of PP polymers during mechanical recycling (Delva et al., 2014), also indicated by the MFI results. However, what was more noticeable was that the variations in impact strength between the replicates for the same sample were significant for all samples, including the external ones, produced at large industrial-scale facilities. This represents a considerable challenge in terms of applicability, as such variation most likely will make the reprocessed PP unsuitable for applications where impact strength is a crucial property for performance.

Moreover, the results indicated that contamination from other polymers, especially multilayer sealing foil, reduced both tensile properties and impact strengths of the reprocessed PP. This is based on the observation that meat trays, where the foil was removed (*PP.Meat.NoFoil*), exhibited considerably higher mechanical properties than when the foil was left on (*PP.Meat.DS* and *PP.Meat.All*). Thus, the chemical heterogeneity of samples including sealing foil, as demonstrated by the TGA results, seems to influence mechanical properties negatively. This is supported by Klitkou et al. (2011), who reported that especially the impact strength of PP is influenced negatively by aggregates of contamination not effectively distributed in the sample.

3.4. Closing the loop for individual polymers and product types

The results revealed that the three polymers, PET, PE and PP, exhibit different properties and therefore act differently during recycling. PET is theoretically well-suited for recycling multiple times, as the degradation in all product types can be reversed during decontamination, if the recycling process is designed accordingly. The importance of a homogenous waste stream is thus less important. On the other hand, the results showed considerable variation regarding processability for different packaging types, such as trays, bottles and tubs, for PE and particularly PP, indicating that the homogeneity of these waste streams is crucial for closed-loop recycling into new packaging products. Consequently, separate management of PP food packaging, as suggested by Eriksen and Astrup (2019), is in itself not sufficient to ensure closed-loop recycling, as PP waste has a high degree of heterogeneity, even within food packaging (Eriksen and Astrup, 2019). Hence, solutions contributing to an increase in the homogeneity of the PE and PP plastic waste sent to reprocessing are crucial, for ensuring conditions enabling closed-loop recycling. Selected strategies are discussed below.

3.4.1. Additional sorting

A potential technical solution includes additional sorting of waste prior to reprocessing. In current recycling practices, plastic can be sorted mechanically based on their polymer type (using

near-infrared spectrometry (NIR)) or colour (using optical sorters) (Turner, 2018; Brunner et al., 2015). These technologies separate the plastic waste based on intrinsic properties. However, they cannot separate different grades of plastic containing the same polymer (Gao et al., 2017), e.g. PP with a high MFI from PP with a low MFI. Consequently, several studies have focused on developing tracer-based sorting approaches, where a tracer (based, for instance, on fluorescence) is added to specific plastic types or products in the production phase, enabling later identification during sorting (Brunner et al., 2015). Although several technical limitations are still associated with such sorting approaches, e.g. tracers made from rare earth elements (Brunner et al., 2015; Gao et al., 2017) involving environmental impacts and potential food safety challenges, they nevertheless represent a unique potential for mechanical separation of products with different material properties.

3.4.2. Regulatory harmonisation

Another type of solution involves regulatory measures aimed at controlling the composition of plastic waste generated in households through harmonisation of product design. This could involve measures requiring all PP trays for food contact purposes to be produced using the same processing method, thereby involving plastic with a similar processability. Combined with measures requiring all food contact bottles to be produced in PET, it could facilitate closed-loop plastic recycling of PET food packaging and PP food trays, simply based on existing NIR sorting technology, if all non-food packaging were simultaneously produced in other polymers, for example PE. As part of the EU's plastic strategy, the Commission announced that they would revise the essential requirements for placing plastic on the market (described in directive 94/62/EC on Packaging and Packaging Waste), to ensure that all plastic packaging is reusable or easily recyclable in 2030, emphasising that product design needs to be addressed more systematically (EC, 2018). Harmonising product design, as suggested here, fits well into this aim and would have the most optimal effect if implemented in a European, rather than a national, setting.

3.4.3. Limiting the effect of PP degradation

Even if the degree of PP waste homogeneity could be increased significantly, PP is suitable for recycling only a limited number of times, due to degradation during recycling. Since PP represents a significant share of plastic waste from households (e.g. Eriksen and Astrup, 2019; van Velzen, 2013), this limits the overall recyclability of PP and thus plastic from HHW in general.

One way to improve recyclability could be to limit the decrease of MFI during recycling through the addition of suitable additives, which Kozłowski (2015) demonstrated is possible. Another way could be to limit the use of PP in short lived packaging applications and instead apply it in long-lived products, such as car parts, building materials, etc., so the PP material is kept in the loop longer. The final option could be to recycle the plastic chemically instead of mechanically, thereby breaking it down into its individual constituents, from where new plastic of food-grade quality can be produced (Ragaert et al., 2017).

Critical limitations, however, might potentially be related to all options; for example, the addition of additives stabilising the MFI might be a challenge for food safety. Moreover, limiting the use of PP in packaging applications might increase the heterogeneity of the PET and PE waste. Even though this might not be a problem

Fig. 4. Visual presentation of tensile strength and strain (at break for PET samples, at yield for PE and PP samples) and impact strength for all samples. PET samples were processed without a dedicated drying process, and are not expected to represent PET recycled at industrial scale. Precise values are provided in Appendix B3 in the SM. nb. = no break.

for PET recycling, it might make the PE waste unsuitable for closed-loop bottle to bottle recycling, if not complemented with additional solutions. Finally, chemical recycling is currently an economically unsound solution (Ragaert et al., 2017) that might not be the most preferable environmental option (Faraca et al., 2019).

In summary, additional mechanical sorting and better regulatory harmonisation of plastic packaging design represent solutions with important potentials to mitigate the many challenges associated with the large heterogeneity in plastic waste. Further quantitative assessments are necessary to validate the effect of concrete solutions and to assess potential trade-offs.

4. Conclusion

Samples of reprocessed PET, PE and PP from households were prepared from waste as well as collected from industrial reprocessing facilities. The samples were analysed with respect to thermal degradation, processability and mechanical properties with the aim of evaluating material quality and suitability for closed-loop recycling.

The study highlights that PET plastic is theoretically well-suited for closed-loop recycling, as potential polymer degradation can be reversed in a decontamination process in order to meet bottle and food-grade PET quality. However, the results showed that moisture control is a key requirement when converting reprocessed PET into products. The reprocessed PE samples from Danish source-separated plastic showed high tensile strength compared to virgin PE, and the processability suggested that closed-loop recycling of non-food bottles was feasible, both when recycled separately and as mixed PE waste. Polymer cross-contamination from lids and labels had little influence on PE quality. The mechanical properties of reprocessed PP were slightly reduced compared to virgin PP, with multilayer sealing foils leading to further reductions. Large variations in impact strength were observed for the individual PP samples, making them unsuitable in applications where impact strength is important. Moreover, the processability of different PP packaging types in the waste varied significantly, indicating that recycling of mixed PP waste and even individual waste packaging types, such as fruit and vegetable trays, cannot facilitate closed-loop recycling into new packaging products. Thus, the heterogeneity of PP waste, even if food-packaging is managed separately, as well as polymer degradation during recycling represent crucial limitations for PP waste recycling.

Considerable differences were demonstrated for PET, PE and PP recycling, which are crucial for the design of plastic recycling systems. The results highlight the importance of homogeneity in PE and PP waste streams with respect to the material properties of the packaging types in the waste. The necessary degree of homogeneity may be reached by additional, tracer-based sorting of the plastic waste prior to reprocessing, regulatory harmonisation of plastic product design, or chemical recycling. Future research should quantitatively assess the concrete effects of such initiatives.

Declaration of Competing Interest

None.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.wasman.2019.07.005>.

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