

RESEARCH ARTICLE

The impact of impurities on the mechanical properties of recycled polyethylene

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Abstract

The mechanical properties of mechanically recycled polyethylene (rPE) were studied in relation to the composition of the feedstock. This composition varied in six steps from only bottle bodies from a single type of high-density polyethylene (HDPE) to the complete composition of an industrially sorted polyethylene (PE) product, including other packaging components, other PE-based packages, sorting faults and residual waste. The rPE with the highest impact resistance was made from the single-grade bottle bodies. The addition of bodies made from other types of PE already reduced the impact resistance with 11%. All the other stepwise additions of packaging components and faultily sorted objects caused the impact resistance to decrease further. Conversely, the elongation at break grew with the stepwise addition of these packaging components and faultily sorted objects. From the used methods, the best analysis method to accurately determine the polymeric composition of the rPE was found to be near-infrared-assisted flake analysis. This method can not only be used to determine the polymeric composition, but due to the strong correlation with the impact resistance, it is also a valuable indicator for the expected mechanical properties of rPE.

KEYWORDS

mechanical properties, mechanical recycling, polyethylene bottles, polymeric composition, post-consumer plastic waste

1 | INTRODUCTION

The European Union strives towards a circular economy to enhance resource efficiency and reduce waste generation.¹ Plastic packaging materials are one of the five policy focus areas, because these have a relative short life span and are dominantly present in waste streams.

Producers of fast-moving consumer goods (FMCG) are simultaneously seeking recycled content for their plastic packages to reduce their environmental impact.² However, these companies can often not procure recycled plastics in the qualities that precisely match the virgin plastics they need to replace. The quality aspects that are reported to be critical vary strongly from mechanical properties (impact and

Abbreviations: CNI, Charpy notched impact; DKR, Deutsche Gesellschaft für Kreislaufwirtschaft und Recycling; DSC, differential scanning calorimetry; DSD, Duales System Deutschland; EAB, elongation at break; FMCG, fast-moving consumer goods; FTIR, Fourier transform infrared; HDPE, high-density polyethylene; LDPE, low-density polyethylene; LWP, lightweight packaging; MFI, melt flow index; NIR, near-infrared; PC, post-consumer; PE, polyethylene; PET, polyethylene terephthalate; PMD, plastics, metal packages and beverage cartons; PP, polypropylene; PS, polystyrene; PVC, polyvinyl chloride; rPE, recycled polyethylene.

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tensile strength), optical properties (haze, gloss and colour), processing parameters such as melt flow index (MFI), odour and food safety status. Additionally, virgin plastics are relatively inexpensive and recycled plastics are often considered a liability risk.

Therefore, it is of great relevance to understand the qualities of recycled plastics that can be obtained with conventional mechanical recycling techniques from the post-consumer (PC) plastic packaging wastes that are currently being collected and sorted. Ideally, we need to understand the factors that determine the quality aspects of these recycled plastics. This knowledge will help to further develop mechanical recycling techniques and to define evidence-based design guidelines. Both will help to progress towards a more circular economy for plastic packages. Hitherto, multiple organisations have released design guidelines for plastic packages.^{3–6} These design guidelines were based on the industrial experience of sorting and recycling companies and on the general knowledge of virgin polymer blends. Although valuable, decisive details of these guidelines currently lack a publicly verifiable scientific basis. Additionally, the various design guidelines do oppose each other with respect to important details, which hampers the broad acceptance by the FMCG industry.

The scope of this study is purposely limited to recycled polyethylene (rPE), because it is the most homogeneous sorted product from PC collection systems in the Netherlands, being predominantly composed of bottles.^{7,8} Although high-density polyethylene (HDPE) bottles prevail in the feedstock, some low-density polyethylene (LDPE) bottles and other types of polyethylene (PE) are also present; therefore, this recycled plastic is named rPE and not, for instance, recycled HDPE.

Hitherto, most technological studies on the recycling of PE packaging waste dealt with either post-industrial rPE,^{9,10} blends of virgin plastics with various rPE^{11–13} and the repetitive processing of virgin PE.^{14,15} Only a limited amount of studies analysed rPE made from sorted products that originate from PC plastic packaging waste collection and sorting schemes.^{16–20} The tensile strength and impact resistance of PC rPE are usually reduced in comparison with virgin HDPE. Conversely, the elongation at break of PC rPE is usually substantially larger than of virgin HDPE. But the reported values vary substantially between different samples of PC rPE.

Review articles underline that there are three main quality deterioration pathways for recycled plastics: degradation, contamination with other polymers and particles and molecular contamination.^{21–23} Because most packaging plastics have a relatively short life span, degradation is usually less relevant for PC packaging plastics and contamination with other polymers, particles and molecules prevails. For mechanically recycled PC PE, the most relevant polymeric contaminant is polypropylene (PP) followed by small amounts of polystyrene (PS), polyethylene terephthalate (PET) and polyvinyl chloride (PVC).^{7,8,17} These polymeric contaminants originate both from sorting faults and packaging components.^{7,8} The polymeric contaminants in the rPE are immiscible and typically form blends, of which the mechanical properties are often reduced as compared with pure PEs.²³ Although the average concentrations of polymeric contaminants in Dutch PC recycled plastics are known,^{7,8} as well as their origins, their impact on the properties of recycled plastics can

only partially be explained by the formation of blends. Furthermore, the packaging components supply not only polymeric contaminants to the recycled plastic but also other types of contaminants, such as glue residues, printing ink residues and additives, which are not removed during the washing process. These other contaminants will potentially also affect the properties of the recycled plastic. With respect to the molecular contaminants present in rPE, significant progress has been made in recent years to understand the types of molecules present and their origin.^{24,25} Four different origins of molecular contaminants are discerned—polymer degradation products, migrated substances, fragrances and microbiological metabolites—of which the latter two are strong odourants that could not be removed with standard mechanical recycling technologies.²⁶

This study has two main objectives. First is to quantify the impact of sorting mistakes and packaging components on the mechanical properties of the rPE in a systematic manner, and second is to develop a test method to assess the purity of rPE (PE content) and predict its applicability. As a consequence, this study contributes to the circular economy for plastic packages by providing a quantitative scientific basis for design-for-recycling guidelines.

2 | MATERIALS AND METHODS

Samples of sorted PE products (36, 82 and 211 kg) were taken at three different sorting facilities in 2014 and stored at 7°C. One sorted PE product originated from the Belgian separate collection system of plastic bottle, metal package and beverage carton (PMD) material, operated by Fost Plus. Another sorted PE product originated from the Dutch separate collection system for PC plastic packages, operated by Nedvang. The last sorted product originated from the German separate collection system for lightweight packages (LWPs: plastic packages, metal packages and beverage cartons) operated by Duales System Deutschland (DSD). The German and Dutch sorted PE products were produced to comply to specification Deutsche Gesellschaft für Kreislaufwirtschaft und Recycling (DKR) 329.²⁷ For this study, all three samples were manually object-wise sorted into 50 different categories of plastic packages and residual materials with a SIRO IOSYS near-infrared (NIR) machine²⁸ (see supporting information S2). The samples were reconstituted and mechanically recycled in our laboratory as previously described.²⁹

Additionally, the Dutch sample of sorted PE was used to prepare six different types of PE feedstock for mechanical recycling with an increasing level of impurities, ranging from the non-coloured, translucent HDPE milk bottle bodies only (code 1) to the sorted PE product as it was received, complete with non-PE packaging components, sorting faults and residual waste (code 6) (see Table 1). This milk bottle is ubiquitously present in Dutch sorted PE. All samples of PE feedstock were mechanically recycled as previously described.²⁹ The washing was carried out at 50°C with 0.01-M sodium hydroxide as default. The most pure feedstock (1) and the most contaminated feedstock (6) were recycled in two compositionally identical batches A and B. Additionally, two batches of the most contaminated

TABLE 1 The six different types of PE feedstock that were mechanically recycled (50°C 0.01-M NaOH), starting with the single HDPE bottle bodies and gradually increasing the complexity to the PE sorting product according to DKR 329 by adding different packaging components and objects

Code	Feedstock composition
1AB	Only non-coloured, translucent HDPE milk bottle bodies
2	PE bottle bodies (all colours and all types)
3	Complete PE bottles and PE flasks including packaging components made from non-PE polymers such as labels, caps and closures
4	Complete PE packages, including, bottles, flasks, rigids and PE films
5	PE packages including faulty sorted objects from predominantly PP and PET
6AB	DKR 329, industrially sorted PE including faulty sorted objects and attached residual waste
6CD	DKR 329, industrially sorted PE including faulty sorted objects and attached residual waste, recycled with cold 0.01-M NaOH solution
6E	Industrially sorted PE from the Belgian Fost Plus system
6F	Industrially sorted PE from the German DSD system

Abbreviations: DKR, Deutsche Gesellschaft für Kreislaufwirtschaft und Recycling; DSD, Duales System Deutschland; HDPE, high-density polyethylene; PE, polyethylene; PET, polyethylene terephthalate; PP, polypropylene.

feedstock (6) were mechanically recycled with cold (11°C) 0.01-M sodium hydroxide solution C and D. The floating products from the sink–float separation vessel were dried overnight in an oven (85°C) and further used for processing.

The dried flakes of floating product were milled with a Wanner C17.26 mill with a 0.8-cm screen and subsequently extruded with a Berstoff ZE 25 (25 cm, 40D) co-rotating twin-screw extruder. The milled dried flakes were fed in the extruder by a gravimetric feeder provided with a nodular screw. To prevent bridge formation, the flex wall from the feeder was switched on. A scheme of the extruder configuration is shown in Figure 1. Z1 to Z10 and Z13 were the standard zones of the extruder where temperatures are set. Z14 was the zone between the melt filter and the die. The pressure in the extruder can rise in case the filter is blocked. Pressure before the filter (Z13) was

monitored during the process. Set point temperatures used during extrusion are also given in Figure 1.

The extruded strands were water-cooled and granulated. These granulates were dried in a Gerco desiccant air dryer for 4 h at 80°C and converted into test specimens in the shape of dog bones according to ISO 527-2 and impact test bars according to ISO-294-1 with a DEMAG D25NC IV injection moulding machine. The barrel temperature was 220°C, and all materials were injected into a cold mould at 30°C.

All specimens produced via injection moulding were conditioned during a week in a controlled environment at 20°C and 50% relative humidity (RH). The mechanical properties were determined according to ISO 527-1 with a Zwick Z010 universal testing machine provided with Multisens extensometers. The E-modulus was determined at a speed of 1 mm/min and the maximal force and elongation at break at 10 mm/min. Charpy notched impact properties were measured according to ISO 179/1eU using a Ceast Resil 50 pendulum impact tester.

The Fourier transform infrared (FTIR) spectra were measured with a Varian Scimitar 1000 FTIR in attenuated total reflection (ATR) mode. Four spectra were taken per sample at a resolution of 4 cm⁻¹. For each spectrum, 45 scans were executed in the range of 650 to 4000 cm⁻¹. The PP content of the recycled plastics was estimated using normalised maxima. The PP absorption at 1376 cm⁻¹ was normalised to the PE absorption at 2848 cm⁻¹ and used to calculate the PP content in fourfold and averaged.¹⁷ The PE content of the samples could not be estimated in a reliable manner from the infrared (IR) data because all PE peaks overlap with PP peaks.

The thermal properties of the recycled products were studied with a Perkin Elmer Diamond differential scanning calorimetry (DSC) provided with auto sampler. The heating rate was 10°C/min, and reported values were those of the second heating scan after quenching. The melting peaks near 120°C with a melting enthalpy of 186 J.g⁻¹ and 160°C with a melting enthalpy of 207 J.g⁻¹ were used to estimate the PE and PP content, respectively.

The MFI of the samples was determined in accordance with ISO-1133 in a Zwick & Roell MFLOW machine at a standard weight of 2.16 kg at a temperature of 240°C. Measurements were performed in sixfold and averaged.

The polymeric composition of the dried flake samples 1A to 6D was determined using an IOSYS SIRO NIR by keeping every single flake with metal tweezers in the light beam for identification and

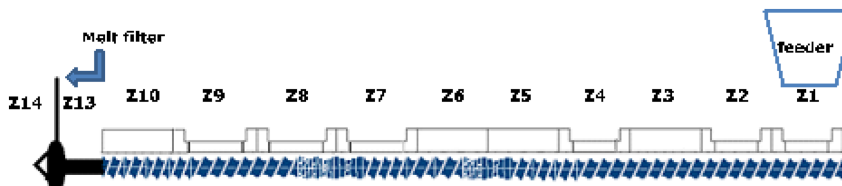


FIGURE 1 Scheme of the extruder configuration used for processing the recycled materials and the temperature settings per zone used for the extrusion of the polyethylene samples when a melt filter was used

Zones	Z14	Z13	Z10	Z9	Z8	Z7	Z6	Z5	Z4	Z3	Z2	Z1
Temperature (°C)	250	250	220	220	225	220	220	220	215	215	90	-

sorting 500-g samples. After sorting, the masses of the various polymer types were weighted, from which the polymer composition was derived. The polymeric composition of the flake samples 6E and 6F was measured with a SiroPAD analyser and a dedicated protocol for rigid samples.²⁰

The colour of the pellets was determined with a Konica Minolta Chroma-meter CR-400. The pellets were spread out in a 10-cm diameter Petri dish and measurements were conducted at five different locations. The resulting CIELAB values (L^* , a^* , b^*) were averaged. These colour parameters were combined into an overall colour change parameter: ΔE . It describes the total colour difference between the rPE made from only milk bottle bodies (averaged values of samples 1A and 1B) as compared with the rPE made from other samples: $\Delta E = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$.

3 | RESULTS

3.1 | Mechanical recycling yields

The mechanical recycling yields of the feedstock types 1A–6D have previously been reported,²⁹ and the recycling yields of samples 6E and 6F are listed in Table S1. The mechanical recycling yields for the three sorted PE products from three different countries and collection systems are largely comparable. The recovery of mass of the floating product was 95–96% for samples 6A–D, 94% for sample 6E and 93% for 6F. The only noticeable difference is that the Belgian sorted PE (6E) yields less sinking by-product in comparison with the Dutch and German sorted PEs (6AB and 6F).

3.2 | Processing of the rPE

The process characteristics of the extrusion of the milled dried flakes to granulates are listed in Table 2. Two process parameters are

reported: the fluctuations in the feed supply rate as compared with the average feed supply rate of 6 kg.hr⁻¹ and the pressure increase in the extruder in front of the 50- μ m melt filter.

The dried flakes of the floating products made from feedstock 1 to 4 could easily be extruded; this is reflected in the low pressure increase in the extruder just before the melt filter. Extrusion of samples 5 and 6A–6F was more complicated. First of all, the supply rate of flakes was more variable, especially for sample 5, which was relatively the richest in plastic film flakes and these are well known to show bridging behaviour in cone-like feeding hoppers. On top of that, the pressure before the melt filter increased strongly for these samples. This can be attributed to both the presence of other polymers such as PET with higher melting points (in samples 5 and 6) and the presence of residual waste (in samples 6A–6F) such as non-melting paper fibres, textile fibres and metallic flakes, which can clog the melt filter and raise the pressure in the extruder. This shows that the processing of rPE made from sorted PE products with a standard mechanical recycling process is not straightforward and certainly different to the processing of virgin materials.

3.3 | Characterisation of the rPEs

The results of the three types of compositional analysis of the rPE samples are listed in Table 3. Both the NIR and the DSC method show that the amount of PE in rPE decreases from sample 1A to 6A and 6B. Conversely, all three methods reveal that the amount of PP in rPE increases between those samples. Furthermore, the Belgian rPE sample (6E) had a significantly higher PE content than the Dutch (6A–D) and the German rPE (6F) samples, which was expected based on the composition of the feedstock (see supporting information S2).

The results of the MFI are listed in Table 2. The rheological measurements indicate that the rPEs are viscous materials with MFI below 1 g/10 min. The rPE made from a single grade of PE (1AB) has a high

TABLE 2 Two relevant extrusion process parameters and the melt flow index measured on the extruded pellets

Code	Fluctuations in feed rate (%)	Pressure increase in the extruder in front of the melt filter (bar)	MFI of extruded pellets, (g/10 min at 240°C and 2.16 kg)
1A	7	20	0.84 ± 0.01
1B	17	16	0.84 ± 0.01
2	4	22	1.11 ± 0.02
3	8	27	0.69 ± 0.01
4	20	23	0.86 ± 0.01
5	58	97	1.21 ± 0.03
6A	17	56	1.33 ± 0.04
6B	11	81	1.20 ± 0.07
6C	23	57	1.22 ± 0.02
6D	22	72	1.27 ± 0.02
6E	Not registered	57	0.89 ± 0.01
6F	Not registered	71	0.98 ± 0.02

Abbreviation: MFI, melt flow index.

TABLE 3 Polymer composition of the recycled PE flakes with NIR, granulates with DSC and test specimen with FTIR in attenuated total reflection mode (%)

Sample	Flakes with NIR		Granulates with DSC ^a		Test specimen with FTIR
	PE	PP	PE	PP	PP
1A	100	0	100	0	0
1B	100	0	100	0	0
2	100	0	97	0	0
3	97.2 ^a	2.3 ^a	90	1.5	5.3 ± 0.7
4	95.8 ^a	3.9 ^a	88	1.4	5.7 ± 0.6
5	89.9 ± 1.5	8.3 ± 1.5	82	2.1	9.1 ± 0.7
6A	90.0 ± 1.3	8.2 ± 1.0	83	2.0	8.6 ± 0.9
6B	90.0 ± 1.3	8.2 ± 1.0	83	2.3	8.1 ± 1.1
6C	90.0 ± 1.3	8.2 ± 1.0	81	2.6	9.6 ± 0.3
6D	90.0 ± 1.3	8.2 ± 1.0	83	2.3	10.2 ± 0.8
6E	95.8 ± 0.0	4.1 ± 0.1	78	1.3	6.1 ± 0.3
6F	89.6 ± 0.9	8.4 ± 0.7	70	2.4	11.0 ± 1.0

Abbreviations: DSC, differential scanning calorimetry; FTIR, Fourier transform infrared; NIR, near-infrared; PE, polyethylene; PP, polypropylene.

^aSingle measurement, hence no error.

viscosity, but the rPE made from a mixture of PE grades has lower viscosity (higher MFI sample 2). When polymeric contaminants are added from labels, caps and closures (sample 3) and from other type of PE-based packages (sample 4), the viscosity increases again. But when faultily sorted objects and residual waste items are added in samples 5 and 6A–D, the viscosity lowers. The variations in the viscosity between the samples probably reflect the MFI values of the contaminants added and their complex influence on the MFI of the PE matrix.

The results of the colour measurements on the test specimen are listed in Table S4. The L* lightness parameter clearly reduces from sample 1AB to 6AD meaning that the pellets become darker coloured. Consequently, the overall change in colour parameters (ΔE) increases between the same samples.

3.4 | Mechanical properties of the rPEs

The mechanical properties of the rPE samples show limited variation (see Table 4). The stiffness (elastic modulus) of the rPE is slightly higher for the most pure samples 1A and 1B and for the Belgian sample 6E and slightly lower and fairly constant for all other samples. The maximum strength of the material is also fairly constant. This was expected, because these properties relate mostly to the bulk of the material and these rPE samples have polymeric purities of 90% and more. The elongation at break and the impact resistance did, however, show clear correlations with the polymeric purity of the rPE samples (see Section 3.5).

The relative high elongation at break of samples 5, 6A–6D and 6F in comparison with samples 1A–4 and 6E most likely relates to the

TABLE 4 Mechanical properties of the recycled PE samples

Code	Elastic modulus (MPa)	Maximal force (MPa)	Elongation at break (%)	Charpy notched impact resistance ($\text{kJ}\cdot\text{m}^{-2}$)
1A	1020 ± 30	26 ± 2	270 ± 160	27.0 ± 1.1
1B	1010 ± 20	24.6 ± 0.6	180 ± 100	28.7 ± 1.2
2	910 ± 30	22.8 ± 0.6	200 ± 100	24.5 ± 1.3
3	930 ± 20	24 ± 2	290 ± 120	21.4 ± 1.3
4	870 ± 20	23.4 ± 0.8	370 ± 100	18.3 ± 0.6
5	880 ± 13	27 ± 2	520 ± 20	14.2 ± 0.5
6A	900 ± 14	25.1 ± 0.7	512 ± 14	12.2 ± 0.5
6B	890 ± 17	26.6 ± 1.2	535 ± 14	12.5 ± 0.6
6C	900 ± 11	26.6 ± 1.2	535 ± 20	14.1 ± 0.8
6D	890 ± 10	29.2 ± 0.4	571 ± 8	13.0 ± 1.0
6E	1072 ± 23	26.9 ± 0.2	107 ± 10	17.8 ± 1.0
6F	931 ± 59	25.2 ± 0.3	361 ± 48	13.6 ± 1.0

presence of flexible LDPE in the former samples and the absence in the latter samples.

The difference in elongation at break and impact resistance for the rPEs with increasing degree of complexity in the composition has practical relevance for its applications. Although the increase in elongation at break will imply that the rPEs will not fail directly but rather deform, the decrease in impact resistance will imply that the rPEs are more brittle and will fail faster when dropped. For many circular applications of rPE, the impact resistance will be critical and hence limit its applicability. When the rPE samples 1A, 1B and 2 are compared, already a clear and significant decrease in impact resistance is observed. Because the difference between these samples is that samples 1A and 1B are composed of one grade of HDPE used from milk bottles and sample 2 is composed of a mixture of PE grades used for blowing bottles with different colour additives than only transparent, this implies that the mixing of PE grades and colour additives already causes a reduction in impact resistance. When the impact resistance of the rPE samples are further compared, it is noticed that with every additional amount of polymeric contaminants from packaging components (sample 3), from sorting faults and from attached dirt (samples 4 and 5), the impact resistance reduces further. The lowest values are observed for samples 5 and 6 with exception of sample 6E that relate to the relative pure Belgian PE sample.

3.5 | Correlation between the polymeric composition and the mechanical properties

Correlations between the compositional data (Table 3), mechanical properties (Table 4), the MFI (Table 2) and the colour parameters (Table S4) were sought with linear regression analysis (see Table S5). The best linear relationship was found between the Charpy notched

impact and the PP concentration in rPE as determined with NIR flake analysis. An inverse linear correlation was found with a coefficient of determination r^2 of 0.95 (see Figure 2). Closer examination reveals that a slightly improved correlation can be found with a second-order polynomial function with a coefficient of determination r^2 of 0.97 (see supporting information S5 and Figure 2). Because blends of polymers often exhibit antagonistic mixing behaviour, their property–composition diagrams tend to be U-shaped over the complete concentration range.^{23,30,31} Because in this study, rPEs with limited concentrations of PP (up to 10%) are investigated, only the outer left part of the U-shaped figure is observed, which can both be described with a linear and a second-order polynomial function.

Multivariate statistical analysis showed that the best function to describe the Charpy notched impact is a second-order polynomial function with the PP concentration in the rPE according to NIR flake analysis and a linear function with the MFI (see supporting information S5). This function with two variables has a coefficient of determination r^2 of 0.98. The minor role of the MFI is to also bring the PE grade into the equation, as the MFI coarsely relates to the types of PE molecules present in the rPE.

Additionally, a good correlation between the elongation at break and the PP concentration in the rPE according to NIR flake analysis could also be found for only the Dutch samples (1–6D). The coefficient of determination r^2 was 0.97. The Belgian and German samples (6E and 6F) were clear outliers.

Both impact properties and elongation at break have previously been reported to correlate with both the composition of binary virgin HDPE: PP blends^{30,31} and with the thermal mechanical degradation of polymers.^{23,32} This study proves that these properties also correlate with the composition of rPEs with different levels of PP as polymeric contaminant. As the concentration of the secondary polymer (PP) rises, the interfacial surface area between the PE and PP phases

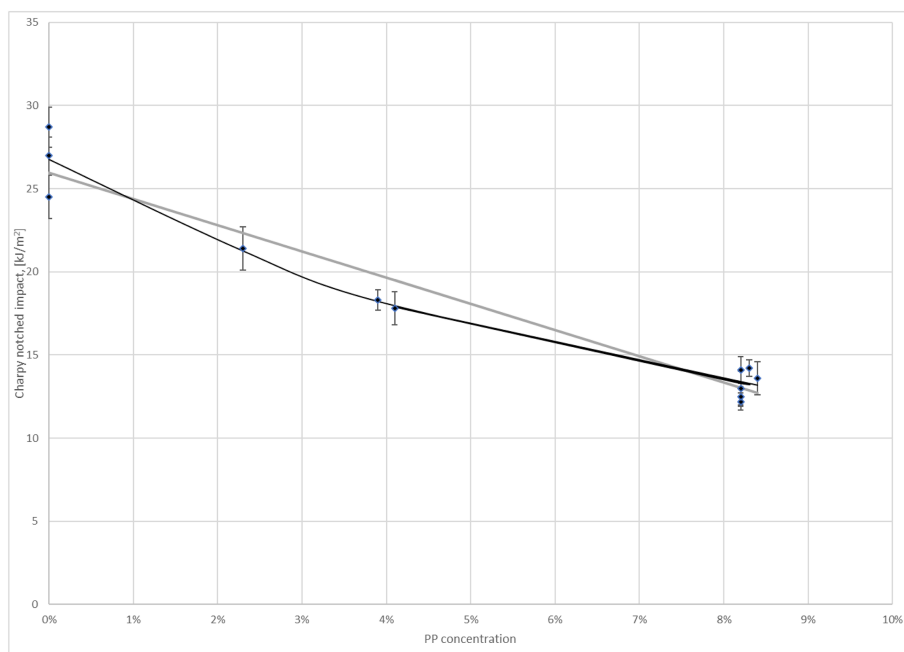


FIGURE 2 The inverse linear relation between the Charpy notched impact and the polypropylene concentration in the recycled polyethylene samples according to near-infrared flake analysis (grey line) and the second-order polynomial relationship (black line)

risers, which results in a less strong material with hence a lower impact resistance. Conversely, the elongation at break increases with the polymeric contamination. The pure recycled material (samples 1A and 1B) is a fairly hard and brittle material, and the presence of PP, and potentially also of LDPE in samples 5, 6A–D and 6F, makes this material more ductile. The fact that the German and Belgian PE samples are outliers probably relates to other grades of PE being used in bottles in these countries and potentially also other packaging components. This suggests that the elongation at break is not only influenced by the PP concentration in the rPE but also by the grade of main HDPE material and potentially also by the presence of other (polymeric and molecular) impurities. This makes this property less useful as a general indicator to assess the quality of a rPE. Additionally, the standard deviations in elongation at break measurements tend to be substantially larger than those in the impact resistance measurement, making the latter more suited as a general quality indicator.

The Charpy impact correlates better with PP concentration as determined by the NIR flake sorting method than with the PP concentrations derived from DSC and IR analysis (see Table S5), which confirms that the NIR flake analysis method is more accurate.

The lightness parameter (L^*) correlates reasonably well ($r^2 = 0.924$) with PP concentration as determined by the NIR flake sorting method for the range 2–6D and 6F, hence excluding the rPE samples made from non-coloured bottles (samples 1A and 1B) and the Belgian sample (6E).

4 | DISCUSSION

4.1 | Techniques to analyse the polymeric composition of rPEs

Three different techniques to determine the polymeric composition of rPEs were compared: FTIR-ATR, DSC and NIR-based flake analysis. The FTIR-ATR technique uses the surface of finished product made from rPE as a substrate or a single extruded pellet. The data acquisition itself is relatively fast and the data analysis is fairly simple. Multiple spectra on the surface of the object can be taken and these spectra can be averaged. The downside is that the technique is semi-quantitatively and insufficiently reliable when the concentration of the impurity PP is below 2%. Furthermore, the IR technique cannot quantify important contaminants such as PS, PET and PVC in the relevant subpercent concentration range. Finally, this method can be used to estimate the concentration of PP in rPE when the PP concentration exceeds 2%. But the method overestimates the amount of PE in rPE when PP is present due to overlapping peaks of PE and PP.

The DSC technique works with the extruded pellets. It is a simple and fast technique. It gives the thermal transitions of polymers; for semi-crystalline polymers, the melting peak can be used to approximate their composition so for instance PE, PP and PET but not for PS and PVC, which are amorphous polymers. Furthermore, the presence

of impurities and the broad distribution of molecular weights present in recycled products can affect the measured melting enthalpy of the present crystals, and therefore, the derived compositions are rough approximations. As Table 3 clearly shows, the DSC-derived PP concentrations are substantially smaller than the NIR-derived PP concentrations measured on flake level from the same sample. It is therefore likely that DSC underestimates the PP concentration in rPE.

The NIR-based flake analysis is a more reliable method to obtain the polymeric composition of recycled plastic flakes. The data acquisition is laborious and requires a good protocol to minimise NIR identification mistakes.²⁰ The data analysis is straightforward. The main advantage is that it yields a fairly complete description of the composition, including all the polymers, textile fibres, paper fibres, metal pieces and glass pieces, with as exceptions the black, dark-coloured polymers and multilayered materials. The major downside is that washed milled goods have a substantial variation in composition, as is for instance clear in the difference in results between samples 5, 6A and 6B. Therefore, it would be wise to take several flake samples and average the compositional data. Additionally, the polymer composition of dried flakes is not necessarily the same as of the extruded pellets, because the melt filter can selectively retain polymers with higher melting points. In short, all studied analysis techniques to study the polymeric composition of recycled plastics have pros and cons, operate on different samples, require more or less effort and render results with different levels of accuracy. Nevertheless, the NIR-based flake analysis yields the most accurate polymeric compositions of the recycled plastic.

This NIR-based flake analysis technique is also best suited to assess the quality and applicability of rPEs, due to its good correlation with the impact resistance measured in samples as prepared in this study, with the use of a melt filter. The most pure rPEs (>98% pure) are suited for recycling in non-food bottles, whereas the less pure rPEs are more suitable for cable lining, drainage pipes and so forth.

4.2 | Factors impacting the quality of rPE

In this study, the mechanical properties of rPEs are studied in a systematic manner in relation to the feedstock composition: from the core bottle bodies to the complete sorted product. The complexity of the feedstock is increased with every sample, directly influencing the composition of the rPE and its properties.

As it is apparent from Table 5, all changes in the complexity of the feedstock result in a further lowering of the Charpy notched impact. The first step (between sample 1AB and 2), in which the feedstock changes from a mono-grade HDPE material to a mixture of various HDPE polymers with colour additives, already results in a 12% drop in the impact strength. When the concomitant packaging components (labels and caps) are added, the impact strength drops a further 11% (from sample 2 to 3). When the feedstock is then further expanded to also contain non-bottle PE packages as they occur in the sorted product, the impact strength drops a further 11% (from sample 3 to 4). When the sorting faults are also added as they occur in the

TABLE 5 Relation between the changes in feedstock composition and the relative impact on the Charpy notched impact

Steps	Change in sample composition	Relative impact on the Charpy notched impact (%)
1–2	From one type of body to a mixture of PE bodies	–12
2–3	From only bottle bodies to complete PE bottles, hence adding packaging components as labels and caps	–11
3–4	From complete PE bottles to a mixture of all PE packages as they occur in the sorted product, hence adding non-targeted PE packages	–11
4–5	From only PE packages to all plastic packages as they occur in the sorted product, hence adding the sorting faults	–15
5–6	From only plastic packages to the complete sorted product, hence adding the residual waste components	–7

Abbreviation: PE, polyethylene.

sorted product, the impact strength lowers with 15% (from sample 4 to 5), and when the residual waste components are also added, the impact strength lowers with another 7%.

rPE produced from conventional feedstock that complies to sorting specification DKR 329 and produced with conventional mechanical recycling technology has an inferior impact resistance as compared with rPE made from one type of HDPE. The reduction of the impact resistance relates both to the presence of various HDPE grades and polymeric contaminants with the rPE matrix. These polymeric contaminants originate from both sorting faults and packaging components.

These results clearly show that a top-quality rPE with a high impact resistance can only be produced from a feedstock that contains a single grade of PE and no other polymers. The presence of both other grades of PE and of other polymers causes a reduction in the impact strength of the rPE.

4.3 | Towards more circular recycling of PE/implications for design guidelines

A fraction of the rPE is currently used for the production of non-food bottles, which is a clear example of a circular economy for plastic packages. For the use in non-food bottles, the rPE has to have a high stiffness, high impact resistance, a low MFI and should be easy to extrude. For some bottle applications, a high resistance to environmental stress cracking is an additional requirement.^{33,34} Because

impact resistance is coarsely related to the resistance for environmental stress cracking and less laborious to measure, it is used as indicative parameter. The highest quality of rPE in terms of impact resistance can only be obtained by recycling the bodies of one single type of HDPE bottle, since the recycling of a mixture of different HDPE bottle bodies (which are made from various grades of HDPE) already results in a rPE with a lower impact strength. Multiple strategies can be followed to generate a high-quality, mono-grade rPE. The most straightforward strategy is to set up a mono-collection system for a single type of PE bottle, like for instance the British milk jug collection system.^{35,36} The design-for-recycling guidelines will then have to focus on the efficient separability of the packaging components label, cap and closure ring from the body. This translates into using a water-soluble glue for the labels and the use of different polymers for the packaging components than of the body, which can be removed by wind sifting, sink-float separation and NIR flake sorting. Additionally, prints such as the best before date should be printed on the label and not on the bottle body to avoid contamination of the rPE.

An alternative strategy is to co-collect the PE bottles with a LWP collection system and to use advanced sorting technologies on the object level to retrieve only this type of PE bottle from the LWP waste, such as marking technologies.^{37,38} This approach still requires the same design-for-recycling guidelines to be successful.

Another alternative strategy is co-collect the bottles with LWP, sort it conventionally, mechanically recycle the PE conventionally and to use advanced sorting technologies on the flake level, such as logarithmic sorting,³⁹ to obtain a single grade of rPE. This approach will also benefit from the same design-for-recycling guidelines.

The results of this study can be regarded as further support for the existing design-for-recycling guidelines.^{3–6} However, in case circular recycling towards non-food bottles is preferred, then this study revealed that more strict guidelines will be required to attain that objective.

5 | CONCLUSION

A mechanical rPE with a high impact resistance can only be made from a feedstock that is composed of one type of bottle bodies that is composed of a single grade of HDPE. The stepwise addition of other bottle bodies (made from other grades of HDPE), packaging components such as labels and caps, sorting faults and residual waste results in a progressive reduction of the impact resistance. The mechanical rPE that can be made from the sorted product DKR 329 with conventional mechanical recycling technologies consequently has an impact resistance that is less than half of the single-grade feedstock. A more circular economy for PE bottles therefore requires either a mono-collection system or improved sorting technologies.

The most accurate analysis method to determine the polymeric composition of rPE is NIR-assisted flake analysis. This method does not only determine the polymeric composition, due to its strong

correlation with the impact resistance, it is also indicative for the quality of the rPE.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of this article.

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