Comparison of the quality of mechanically recycled plastics made from separately collected and mechanically recovered plastic packaging waste

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Abstract

In the Netherlands plastic packaging waste is collected via separate collection systems that are highly comparable to the German yellow bag (LWP) system and also retrieved via mechanical recovery from the mixed municipal solid waste (MSW). The retrieved plastics are sorted and mechanically recycled in facilities in the Netherlands and Germany. In this study we compare the quality of mechanically recycled plastics made from separately collected (SC) and mechanically recovered (MR) plastic packaging waste. The quality of the recycled plastics is highly comparable in many aspects, although subtle differences can be noticed. The polymeric purity of the mechanically recovered recycled plastic tends to be slightly higher than of the separately collected plastic, whereas the particle contamination of mechanically recovered plastics tends to be higher. Both types of recycled plastics smell differently.

Keywords

Recycled plastics; purity; quality; separate collection; mechanical recovery; lightweight packaging waste; post-consumer plastic packaging waste; odour

1 Introduction

Prior to 2009 only large (>0.75 ltr) poly ethylene terephthalate (PET) bottles for water and soda beverages were collected via deposit refund systems (DRS) in the Netherlands. From then on, all other types of post-consumer plastic packages had to be collected, sorted and recycled. Municipalities had to organise the collection of plastic packaging waste (PPW). Most municipalities introduced various forms of separate collection (drop-off, kerbside with bags and wheelie bins). Three northern provinces (Friesland, Groningen and Drenthe) decided to upgrade their existing central waste sorting facilities to fully fledged material recovery facilities (MRF) and they did not set up separate collection systems for LWP. The collection portfolio was initially limited to only plastic packages (BROUWER ET AL., 2018). From 2015 on the collection portfolio was expanded to include also beverage cartons and metal packages, similar as the lightweight packaging (LWP) collection in Germany (BROUWER ET AL., 2019). Both the separately collected LWP and the two recovered concentrates from the MRF 1) rigid plastic packages and beverage cartons and 2) flexible packaging films were sent to sorting facilities. The sorted products were sent to certified recycling companies (EUCERTPLAST, 2021). In the early years, most of the PPW was sorted in Germany and therefore, it was decided to join the system of DKR specifications (GRUENE PUNKT, 2021). In the rural parts of the Netherlands, the separate collection of LWP with wheelie bins became the dominant collection system, yielding participation rates of nearly 100% (THODEN VAN VELZEN ET AL., 2019). However, in the urban centres in western part of the country only drop-off collection systems could be operated. The participation rates were low and hence the collection rates were also low and the collected material contained too much non-targeted contributions. Consequently, city councils decided to stop with separate collection in selected neighbourhoods and to commence with mechanical recovery. From 2017 to 2019 three new MRF's were established in Alkmaar, Amsterdam and Rotterdam, see table 1. As a consequence we have multiple separate collection systems for PPW, mechanical recovery and combinations of both. The combined annual processing capacity of the MRF's equals almost half of the production of the mixed municipal solid waste (MSW) in the Netherlands. This study compares the quality of the recycled plastics resulting from the Dutch separate collection and mechanical recovery systems.

Table 1 Material recovery facilities in the Netherlands that produce LWP concentrates.

MRF name	Location	Start of operation, [year]	Approximate annual capacity for MSW, [kton/a]
Attero	Groningen	2009	60
Omrin	Heerenveen	2009	210
Attero	Wijster	2011	310
HVC Sortiva	Alkmaar	2017	140
AEB	Amsterdam	2018	300
AVR	Rotterdam	2019	400

1.1 Mechanical recovery process

Dutch MRF's that aim to recover LWP use freshly collected mixed MSW as feedstock. The recovery process typically starts with a bag opener, a wind-sifter, a drum sieve, magnets and Eddy current separators. The lightweight fraction is further sorted with a NIR sorting machine to produce either a flexible packaging film product (DKR 310) or even a flexible PE film product. The drum sieve yields three size fractions. The fine fraction (<6 cm) is named organic wet fraction (OWF) and is digested to produce methane, dried and incinerated. The coarse fraction (>25 cm) is manually sorted. The middle sieve fraction is processed by a set of NIR sorting machines, yielding a concentrate of rigid plastic packages and beverage cartons. Which is sent to sorting facilities for further treatment. All MRF's have a slightly different set-up, some have additional ballistic separators, others have vibrating cascade sieves, etc.. Most Dutch MRF's recover 50-60% of the PPW present in the MSW feedstock, the largest losses of PPW (approximately 30%) relate to small plastic objects that end up in the OWF.

1.2 Quality of recycled plastics

Different quality aspects are used to describe the quality of recycled plastics at the three stages of the recycling chain; sorted products (bales), flakes (washed milled goods) and granulate (pellets). The quality of sorted products is best described by manual sorting (THODEN VAN VELZEN ET AL., 2018). The sorting results can be used to test compliance to the DKR specifications and to describe the sorted products in terms of the types of objects (plastic packages, non-packaging plastic objects and residual waste) present. Since most plastic packages are composed of multiple polymer and material types, this description in terms of objects is only indicative for the polymer and material composition. Additionally, the level of attached moisture and dirt (LAMD) is regularly measured for main plastic packaging types present in the bales to assess their cleanliness (THODEN VAN VELZEN ET AL., 2018). On the flake level the polymeric composition can be determined with NIR sorting equipment (ALVARADO CHACON ET AL., 2020). And on the level of pellets the recycled plastic can be studied with common technologies used to study polymers: rheology, differential scanning calorimetry (DSC), mechanical properties, infrared spectrophotometry (IR), haze, thermal gravimetry and technologies to study particle contamination, such as Partisol.

There are three main quality decay mechanisms for recycled plastics: degradation (chain scission), contamination with polymers & particles and contamination with absorbed molecules (odours, migration) (VILIPLANA ET AL., 2008). For the short-lived packages the degradation is usually less relevant Polymeric and particle contamination often seriously negatively affects the quality of the recycled plastic. Most polymers are immiscible in each other and a bit of polymeric contamination therefore already causes the formation of a blend, which usually negatively impacts the mechanical and optical properties (RAGAERT ET AL., 2017). Finally, absorbed molecules can render the recycled plastic an odour, which does not always fits the application and can pose food safety risks (HAHLADAKIS ET AL., 2018).

1.3 Objective

This paper offers an overview of the knowledge that has been gathered in the past decade on the quality of recycled plastics produced from separately collected and mechanically recovered post-consumer plastic packaging waste. The focus is on post-consumer recycled plastics originating from the Netherlands and will be compared with data from different countries, in case it is available.

2 Composition of sorted PPW

Hundreds of sorting analysis have been averaged to yield average compositions for sorted products originating from separate collection prior to 2014 (Brouwer et al., 2018 Table C.1) and after 2017 (Brouwer et al., 2019, dataset Table E) and from mechanical recovery prior to 2014 (Brouwer et al., 2018 Table D.1) and after 2017 (Brouwer et al., 2018, dataset Table H). In general, the compositions are fairly similar, the sorted products originating from mechanical recovery have slightly lower concentrations of objects made from non-targeted polymers such as PS and PVC in comparison to those from separate collection, but the LAMD are higher, see Table 2. The lower concentration of non-targeted polymers in recovered & sorted plastics is likely to be caused by the double NIR sorting; first at the MRF and then at the sorting facility.

Table 2 Average levels of attached moisture and dirt from sorted products (SP) originating from separate collection (SC) and mechanical recovery (MR).

SP	SC 2014	MR 2014	SC 2017	MR 2017
PET 328-1	12%	14%	9%	14%
PE 329	15%	11%	5%	11%
PP 324	11%	17%	10%	17%
Film 310	21%	30%	12%	16%
MP 350	14%	35%	8%	35%

The sorted products made from Dutch PCPPW are fairly similar to the German sorted products, but there are subtle differences. Up to 2022 the concentration of small PET beverage bottles is relatively high in Dutch PET 328-1, but after that year it will be substantially lower due to the introduction of a DRS for these bottles. Furthermore, the Dutch use slightly more PP films, due to differences in consumption patterns and this can be noticed in slightly higher concentrations of PP film in sorted Mixed plastics (DKR 350). A detailed comparison between the composition of Dutch, German and Belgian PE DKR 329 is found in (THODEN VAN VELZEN ET AL., 2021).

3 Mechanical recycling yields

The mechanical recycling yields of sorted bales to dried washed milled goods have been published previously (THODEN VAN VELZEN ET AL., 2017 & 2021). The mass yields of the separately collected sorted products are slightly higher than those from mechanical recovery as could be expected based on their lower level of attached moisture and dirt. Furthermore, the mass yields for sludge and dissolved matter are relatively higher for the mechanically recovered plastics.

Table 3 Polymer composition of the washed milled goods %(m/m).

Sample	Targeted polymer	Non-targeted polymers	Other materials
DRS PET bottles	99.3%	0.6%	0.1%
SC PET 328-1	97.2%	2.8%	0.0%
MR PET 328-1	99.4%	0.2%	0.4%
SC PE 329	90.6%	9.3%	0.1%
MR PE 329	94.0%	3.0%	3.0%
SC PP 324	90.6%	9.2%	0.2%
MR PP 324	95.0%	4.2%	0.8%
SC Film 310	76.4%	22.7%	0.9%
MR Film 310	96.8%	2.8%	0.4%
SC Mix 350	63.5%	30.2%	6.3%
MR Mix 350	72.6%	25.6%	1.8%

4 Composition of washed milled goods

The composition of the washed milled goods, produced with a standardised mechanical recycling process, has been determined NIR assisted manual sorting and with a NIR sorting machine (ALVARADO CHACON ET AL., 2020) and is summarised in Table 3. The washed milled goods made from mechanically recovered and sorted products generally contain slightly more targeted polymer, but also more other materials (paper, wood, glass, metals, etc.) than those made from separately collected feedstock (BROUWER ET AL., 2018; THODEN VAN VELZEN ET AL., 2016).

5 Particle contamination and properties of recycled plastics

Recycled plastics that have been extruded and pelletised can be studied with a range of analysis methods. The difference between separately collected and mechanically recovered plastics has been studied in the greatest detail for PET, therefore the results are split in two paragraphs: recycled PET and recycled polyolefins.

5.1 Recycled PET

Recycled PET (rPET) pellets were produced from three feedstocks (DRS, SC and MR) with a standard mechanical recycling process that also involved a post-condensation process (THODEN VAN VELZEN ET AL., 2016). A summary of the most relevant properties is shown in Table 4.

The comparison in properties shows that the rPET products originating from separate collection and mechanical recovery are more contaminated than the rPET from the DRS. However, the qualities of rPET produced in this study are unsuited for many relevant applications due to their dark colour (low L* values), high haze values and high levels of particle contamination. Lower levels of particle contamination, haze and better colour values can be achieved by advanced recycling processes in which for instance the washed milled goods are also subjected to flake sorting (THODEN VAN VELZEN ET AL., 2016). Therefore the quality of the rPET made from all three feedstocks could be improved. The quality of rPET is very sensitive for particle and polymer contamination. The presence of either type of contamination directly results in a grey, hazy material. Hence, to produce top-quality rPET it is vital to use the best feedstock (DRS) and an advanced recycling process. The higher level of particle contamination in separately collected rPET as compared to DRS rPET has also been reported in a German study from 2017 (SNELL ET AL., 2017).

Table 4 Properties and characteristics of rPET pellets produced from three different feedstocks with standard mechanical recycling processes and post-condensation.

Property	PET DRS	PET SC	PET MR
Colour L*	54 ± 3	51.5 ± 1.5	49.4 ± 0.6
Colour a*	-1.9 ± 0.1	-2.9 ± 0.1	2.9 ± 0.1
Colour b*	1.2 ± 0.5	3.5 ± 0.2	8.1 ± 0.4
Haze, [%]	45.1 ± 0.5	87.7 ± 0.6	84.4 ± 0.3
Partisol, [PPTI]	130.570	1.162.175	695.396
IV, [dl/g]	0.94	0.78	0.75
Mn GPC, [g/mole]	35.700	32.500	35.000
Xc DSC, [%]	26 ± 2	27 ± 2	28 ± 2

+ Partisol is the amount of counted particles in dissolved PET in 10000 images.
+ IV is intrinsic viscosity
+Mn is the normalised molecular weight according to gel partitioning chromatography (GPC)
+ Xc is the degree of crystallisation according to the melt peak in the second heating run in differential scanning calorimetry (DSC).

5.2 Recycled polyolefins

Studies in which the properties of recycled PE, PP, Film and MIX are compared with respect to their collection method (SC or MR) are rare (LUIJSTERBURG ET AL., 2014) and in most cases substantial variations in values are reported. Consequently, no clear difference between the quality and properties of the recycled polyolefins from separate collection and mechanical recovery can be discerned. However, these studies have unveiled that three factors affect the mechanical properties: polymer purity, grade purity and additives.

The particle contamination in recycled polyolefins is hardly ever measured, since the methods are laborious, costly and challenging. Therefore, it is more common to measure indirect parameters such as the composition in terms of the main polymer types (PE and PP) with DSC and IR and mechanical properties (LUIJSTERBURG ET AL., 2014). Most recycled polyolefins contain substantial amounts of the other main polymer type. So, a recycled PE regularly contains roughly 10% PP and a recycled PP often contains about 10% PE. The presence of the other main polymer type has substantial impacts on several mechanical properties of the recycled polyolefin; especially on properties such as strain at break and impact strength and less so on tensile strength (LUIJSTERBURG ET AL., 2014; RAGAERT ET AL., 2017; THODEN VAN VELZEN ET AL., 2021).

Additionally, the mechanical properties of recycled polyolefins are affected by the mixing of multiple grades of the main polymer. Post-consumer recycled PE does not only contain approximately 10% PP, but within the 90% of PE several grades of PE are present: HDPE, LDPE, LLDPE, etc. Although most of these grades are miscible, the mixing of these grades does lower the impact strength. Furthermore, by mixing various PE's from various packages, not only different grades are mixed, but also various additives present in the various packages. This will affect the impact as well (THODEN VAN VELZEN ET AL., 2021).

6 Molecular contamination

Plastics absorb various molecules during their usage and recycling, which hamper the application of recycled plastics.

Recycled PET (rPET) is often subjected to post-condensation to restore the molecular weight of the polymer chains and to remove volatiles (WELLE, 2011). Therefore, rPET rarely has an odour and the gas chromatograms only show a limited amount of small peaks. In table 5 the two most clear peaks in the headspace gas chromatogram of rPET are shown for rPET produced with a standard mechanical recycling process and post-condensation, originating either from DRS, SC or MR. Acetaldehyde is a thermal degradation product of PET (WELLE, 2018) and benzene is formed by the presence of chlorine containing contaminants (probably PVC) (THODEN VAN VELZEN ET AL., 2020). Clearly, the recycled PET made from SC or MR bottles and produced with a standard recycling process did yield the undesired contaminant benzene, which could be avoided by recycling these SC and MR bottles with a more advanced recycling process (THODEN VAN VELZEN ET AL., 2016). It should be noted that studies from other European countries have reported other impurities in rPET. For instance a Spanish article describes the presence of bisphenol-A in rPET at low concentrations (DREOLIN ET AL., 2019).

Table 5 Concentration of volatiles in recycled PET originating from DRS, SC and MR (THODEN VAN VELZEN ET AL., 2016).

Origin rPET	Acetaldehyde, [μg/g]	Benzene, [ng/g]
DRS	3.0	0.0
SC	2.9	1.1
MR	3.1	0.9

Recycled polyolefins are often degassed during extrusion and in some cases even gas-stripped, but in general these polymers contain much more volatile compounds. Therefore, headspace gas chromatograms tend to show much more peaks than those of PET, in some cases exceeding 10000. The amount of peaks and the peak areas can be reduced by recycling with hot alkaline water instead of cold water and by degassing during extrusion. However, the specific odour can in most instances not be removed completely (STRANGL ET AL., 2019 & 2021). Research on the molecular contamination of polyolefins has been conducted for HDPE (STRANGL ET AL., 2019), PP (STRANGL ET AL., 2021) and flexible films (DEMETS ET AL., 2020; STRANGL ET AL., 2020), the latter is the preferred research object due to its large surface to volume ratio.

A Spanish research group studied the odour of recycled Film originating from SC and MR. They found that the recycled Film originating from SC smelled earthy and musty, whereas the recycled Film originating from MR smelled cheesy and faecal (CABANES ET AL., 2020). Simultaneously, a similar study was executed in the Netherlands, with recycled PE-flexible films originating from the Dutch SC and MR collection systems. Surprisingly, the results differed slightly from the Spanish results (MAASKANT-REILINK ET AL., 2020). Dutch recycled PE-films made from SC feedstock smells fatty, rancid, fruity and almond like. This odour remains unchanged, even after hot alkaline washing and degassing during extrusion. In contrast, the Dutch recycled PE-films from MR feedstock had a strong burnt odour when it was recycled with cold water and smelled soap-like when it was washed with hot alkaline solutions. This strong burnt odour was not present in the cold washed milled goods, but was strongly present after extrusion. This implies that this strong burnt odour was formed during extrusion. Remarkably it did not form after extruding recycled PE film that was washed with hot alkaline solutions. A possible explanation is, that a reactant present in the PE films from the MRF is efficiently removed by the hot alkaline washing treatment, preventing its further reaction in the extruder. A preliminary hypothesis is that the decomposition of organic waste present in the MSW creates amines and hydrogen sulphide gas in low concentrations. These gases absorb in polyolefin based packages that are also present in the MSW. During the mechanical recycling process in which the PE films are washed with cold water, these molecules remain inside the PE film and during the hot extrusion process these gases will react with the alkene degradation products from the PE film to form alkyl amines and alkyl thiols, which are known to be extremely odour active. These amines and sulphides can, however, effectively be removed from the PE films by washing with hot alkaline solutions, preventing the unwanted reaction to occur (MAASKANT-REILINK ET AL., 2020). Further research is required to test this hypothesis.

7 Discussion

The qualities of the recycled plastics that can be produced from LWP originating from separate collection or mechanical recovery do differ in details. The odour, the polymeric purity and the properties can all be slightly different. But these differences are often relatively small and sometimes even irrelevant.

The debate on the best collection method for LWP is polarised, with strong advocates and opponents for both separate collection and mechanical recovery. Both systems have pros and cons and can be executed correctly and incorrectly. Separate collection of LWP in rural areas is supported by high participation rates and yields high collection rates. In the most urban centres of the Netherlands, however, the separate collection system with drop-off containers for LWP only received low participation rates and collection rates. Consequently, it does make sense to mechanically recover PPW from MSW in these urban centres, as the recovery rates are substantially higher than the collection rates.

Initially (2009-2010) multiple sorting and recycling facilities were not very enthusiastic about the mechanical recovery of PPW, as these materials are more contaminated with dirt and grime than the separately collected PPW. For the sorting companies this translated in a lower mass yield and larger amounts of sorting residues that have to be incinerated. Depending on the contract form, this can result in a financial risk for the management of the sorting facility. After a few years of experience, this is now well-understood by the incumbents and multiple sorting facilities gladly sort recovered concentrates. Furthermore, two sorting facilities in the Netherlands are currently engineered to sort recovered concentrates.

Within the community of recycling facilities the opinions are mixed. Some really appreciate the recovered sorted products, as they are slightly less expensive and have higher shares of targeted polymers. These companies, however, do need to remove more undesired materials and hence produce more waste and wastewater. The recycling facility has to be engineered to handle this feedstock and also deal with larger amounts of process waste. This can form a bottle-neck for some recycling facilities, especially when their wastewater management system has a limited capacity.

For the Dutch extended producer responsibility scheme operator Afvalfonds mechanical recovery is crucial to achieve the ambitious national recycling targets.

8 Conclusion

The mechanical recovery of lightweight packages (LWP) from municipal solid waste is currently performed on a large scale in the Netherlands. The produced packaging waste concentrates are sorted to the same specifications as the separately collected LWP and sold to various recycling facilities. The recycled plastics produced from these feedstocks differ only in details and are used in similar applications. Mechanical recovery is vital for the urban centres in the western part of the country, since the alternative separate collection systems with drop-off containers resulted in too low collection rates.

9 Literature

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