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Abstract. The recycling of packaging waste is an important part of the EU circular economy package, with a political focus on raising the recycling targets for post-consumer plastic packaging waste (PPW). The recycling of PPW involves at least three steps; collection, sorting and mechanical recycling. In contrast to the first two steps, mechanical recycling is poorly documented, as it is considered a free market activity. In order to provide a complete chain description the mechanical recycling yields were determined. The recovery of mass was determined for the main plastic sorting products from both major collection systems: separate collection (SC) and mechanical recovery (MR) from municipal solid waste. This technical assessment was conducted with a laboratory set-up for a standard mechanical recycling process. This analysis showed that there is a substantial sample-to-sample variation in polymeric composition between similar feedstocks and this variation is also observed in recovered masses. Next, the mechanical recycling of polyethylene feedstocks was studied more in depth. Six PE feedstocks with a gradual increasing level of complexity (from only transparent PE bottle bodies to the complete PE sorting product according to DKR 329), were prepared and mechanical recycled with the laboratory set-up. Since the polymeric composition of both the six feedstocks and the six floating milled goods were known, the net PE recycling yield could be calculated. The net PE yields are close to 100% for such a standard recycling process. Additionally, the compositional analysis revealed that contaminants are only partially removed by the standard mechanical recycling process.

Keywords: Recycling, post-consumer plastic packaging waste, washed milled goods, yields.
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INTRODUCTION

The recycling of packaging waste is an important part of the EU circular economy package, with a common EU recycling target of 75% for all packaging wastes by 2030 [1]. Plastic is the second-largest packaging material and achieving high recycling targets is relatively more challenging in comparison to the other materials, due to the compositional complexity of plastic packages. Hence, the political focus is on raising the recycling targets for especially post-consumer plastic packaging waste (PPW). This calls for a scientific technological description of the PPW recycling system. The recycling of post-consumer plastic packaging waste involves at least three different steps; collection, sorting and mechanical recycling. The first two steps are well-documented in several countries, since this a legal obligation and the financial remuneration depends on administrative compliance. The subsequent mechanical recycling step (the conversion of sorted plastic products into washed milled goods) is, however, considered a free market activity and therefore this recycling process step is not publically documented. Furthermore, the mechanical recycling step itself is only poorly described in the scientific literature, for limited combinations of collected and sorted plastic packaging feedstocks and processes [2, 3].

In order to scientifically describe the Dutch recycling system for post-consumer plastic packaging waste, the mechanical recycling yields (the relation between the weight of the feedstock and the weight of the products, the washed milled goods) had to be determined for all five sorted plastic products from both major collection systems: separate collection (SC) and mechanical recovery (MR) from municipal solid waste (MSW) [4]. This was achieved by setting up a dedicated laboratory to mimic standard mechanical recycling processes. The standard mechanical recycling process is a simplification of the industrial process and is composed of a shredder, a washing vessel, a float-sink-separator, centrifuge and an oven. It yields two products (the floating fraction and the sinking fraction) as washed and dried milled goods, a waste-product (dried sludge in 3 size classes) and waste water. The recovery of mass is simply derived from the quotient of the product weights and the feedstock weight (equation 2).

The plastic yield can only be determined in case the concentration of plastic in the feedstock and the products are known (equation 3). However, in the industrial practice these concentrations are unknown. This is, nevertheless, relevant since the sorted plastic packaging products do contain different plastics and materials at two levels. At the object-level, the sorted plastic product contains besides the desired plastic packages also faulty sorted objects and some residual waste. At the packaging-level, almost all plastic packages are composed of multiple components made from different materials. For instance a typical small Dutch PET bottle is composed of a PET body (14.7 g), a PP label (0.4 g) and a PE cap with closure ring (2.0 g). In order to study the plastic yields, the composition of sorted PE feedstock was determined in great detail (object-level and component-level), mechanical recycling was conducted and the composition of the products was studied in great detail by manual NIR-assisted flake analysis. This was conducted in a systematic manner, starting with the composition of sorted PE feedstocks and a stepwise reduction in compositional complexity to the level of clean PE bottle bodies, see table 1.

METHODS

Samples of sorted plastic products (25-50 kg) were taken at sorting facilities and stored at 7ºC. All plastic sorted products were sampled, hence PET (DKR 328-1), PE (DKR 329), PP (DKR 324), Film (DKR 310) and Mixed Plastics (MP, DKR 350). The codes refer to the specifications to which these sorted plastic products need to comply [5]. The samples were manually object-wise sorted into 50 different categories of plastic packages and residual materials with a SIRO IOSYS near infrared (NIR) machine. The samples were reconstituted (with the exception of large metal and mineral objects to protect the equipment). Besides full samples also five partial reconstituted samples were prepared, see table 1. The reconstituted feedstock was milled with a WEIMA WLK-04 shredder and a 2 cm ø screen, yielding dirty milled flakes in the 1-5 cm size range. Samples were taken to determine the dry matter content (dm) and the net material content (nmc). The dry matter content is determined by taking a gross sample of plastic flakes, weighing it, placing it an oven for 12 hours at 105ºC and weighing it again; the ratio of the dry weight over the gross weight is the dry matter content. The net material content is determined by taking a gross sample of plastic flakes, weighing it, washing it and drying it in the same manner. The ratio of the net clean weight over the gross weight is the net material content. 5 kg gross plastic was washed with 150 liter aqueous 0.01 M sodium hydroxide solution at 50ºC for 5 minutes in a large metal stirring vessel. Only for plastic film material the amount of feedstock had to be reduced to 2.5 kg gross. The waste water was sieved over three different screens (2 mm, 1 mm and 0.5 mm), sampled and discarded. The three different types of collected sludge were dried and weighed. The moist washed plastic flakes were transferred into a 3 meter high conical shaped settling tank filled with 200 liter tap water to mimic industrial floatation separation. The floating plastic flakes were scooped up from the top of the tank after 2 minutes and the sinking plastic flakes were collected in the bottom sluice by opening the bottom valve. Both the low density plastic flakes and the high density plastic flakes were separately centrifuged and dried in an oven (85ºC, overnight). Both dried products were weighted. The mass of dissolved matter ($m_{dissolved}^{dm}$) was estimated from the difference in the dry weight of the feedstock and the dried weights of the floating product, sinking product and sludges, see equation 1.

$$m_{dissolved}^{dm} = m_{feedstock}^{gross} * dm - m_{floatingproduct}^{dm} - m_{\sin kingproduct}^{dm} - m_{sludges}^{dm} \quad (1)$$

The recovery of mass (R_m) of a product was calculated from quotient of the dry product weight and the dry feedstock weight, which in turn is derived from the product of the gross feedstock weight and the dry matter content.

$$R_m^{Floatingproduct} = \frac{m_{floatingproduct}^{dm}}{m_{feedstock}^{gross} * dm} \quad (2)$$

The PE yield (η^{PE}) for the six samples in table 1 is calculated from the mass of polyethylene flakes in the product divided by the PE composition of the feedstock expressed in dry weights, see equation 3.

$$\eta^{PE} = \frac{c_{floatingproduct}^{PE} * m_{floatingproduct}^{dm}}{c_{feedstock}^{PE} * m_{feedstock}^{gross} * nmc} \quad (3)$$

TABLE (1). The six PE feedstocks tested, starting with the bare essence (the transparent PE bottle bodies) and gradually increasing the complexity to the PE sorting product according to DKR 329 by adding different contaminants.

Code	Feedstock composition
1	Only transparent PE bottles
2	PE bottles (all colors)
3	Only complete PE bottles and PE flasks including packaging components made from non-PE polymers such as labels, caps and closures
4	Only complete PE packages, hence including PE films
5	PE packages including faulty sorted objects from predominantly PP and PET
6 warm	SC DKR 329, sorted PE including faulty sorted objects and attached residual waste, washed with 50°C 0.01 M NaOH solution
6 cold	SC DKR 329, sorted PE including faulty sorted objects and attached residual waste, washed with cold 0.01 M NaOH solution

The material composition of the feedstocks were calculated from two datasets; sorting analysis of feedstocks on the level of packaging categories and average material compositions of packaging categories. This yielded a composition in terms of 6 polymer types (PE, PP, PET, PS, PVC and black) and 2 residual materials in percentages and hence also the concentration of PE in the feedstock ($c^{PE}_{feedstock}$). The material composition of the produced milled goods were determined for 500 gram samples by NIR-based flake sorting with an IOSYS-SIRO NIR analyser and yielded the concentration of PE in the product ($c^{PE}_{floatingproduct}$).

RESULTS

The measured recovered masses for several different sorted plastic products processed with the simple standard process are listed in table 2. The results for slightly more complex processes involving pre-screening and mixed plastics have been presented previously [6]. The average recovered mass of the main product (PE, PP, film: floating product; PET: sinking product; MP: both) varied between 83 and 94%. Although in general the recovered masses are higher for separately collected and sorted plastics (SC) in comparison to mechanical recovered and sorted plastics (MR), there also appears to be a substantial sample-to-sample variation. This is especially true for the ratio between the recovered masses of the floating and sinking product. For the polyolefinic feedstocks (PE, PP, Film) the sinking fraction is mostly composed from PET, PVC, PS packaging components and polyolefinic packages with large amounts of inorganic fillers. The use of these heavy plastic components and inorganic fillers will vary between packages and therefore, these recovered masses should preferably be used indicatively.

TABLE (2). Recovery of mass for various sorted plastic products as feedstocks for a standard mechanical recycling process.

Feedstock	Dry matter content of the feedstock	Recovered mass of floating product	Recovered mass of sinking product	Sludge waste	Dissolved substances
PET SC	93%	11±1%	83±2%	3±2%	3±2%
PET MR	87%	10±2%	84±2%	3±2%	3±2%
PE SC	95%	94±2%	3±1%	0.8±0.3%	2±2%
PE MR	96%	93±2%	2±2%	1±2%	4±2%
PP SC	99%	90±1%	7±1%	0.7±0.6%	3±1%
PP MR	88%	83±2%	3±2%	6±2	8±2%
Film SC	97%	90±6%	4±4%	0.8±0.6%	6±3%
Film MR	87%	83±15%	4±2%	3±4%	10±9%
MP SC	98%	64±12%	32±11%	0.7±0.8%	5±1%
MP MR	87%	65±4%	28±1%	3.1±0.1%	4±3%

The recovered masses for the six different PE feedstocks with an increasing level of complexity are given in table 3 and the material composition of the feedstock and produced milled goods (floating products) are given in table 4.

TABLE (3). Recovery of mass (R_m) for the 6 different PE feedstocks of table 1.

Feedstock	Dry matter content of the feedstock	Recovered mass of floating product	Recovered mass of sinking product	Sludge waste	Dissolved substances
1	99.1%	98.1%	0.2%	0.5%	1±2%
2	98.5%	98.2%	0.0%	0.3%	1±2%
3	92.3%	97.9%	1.4%	0.9%	0±2%
4	91.0%	95.6%	1.4%	1.0%	2±2%
5	93.5%	94.4%	1.9%	0.9%	3±2%
6 warm	92.2%	96.8%	2.2%	0.9%	0±2%
6 cold	92.2%	95.1%	3.6%	0.9%	0±2%

TABLE (4). The composition of feedstocks (FS) and produced milled goods (MG) of the floating products for the 6 PE samples.

Material	1 & 2		3		4		5		6	
	FS	MG	FS	MG	FS	MG	FS	MG	FS	MG
PE	100.0%	100.0%	97.6%	97.2%	97.7%	95.8%	89.3%	89.9%	88.7%	90.0%
PP			0.1%	2.3%	0.1%	3.9%	6.8%	8.3%	6.7%	8.2%
PS			0.0%	0.1%	0.0%	0.0%	0.3%	0.0%	0.3%	0.2%
PVC			0.0%	0.0%	0.0%	0.0%	0.1%	0.1%	0.1%	0.3%
PET			0.0%	0.2%	0.0%	0.0%	0.8%	0.0%	0.8%	0.2%
Black plastics			0.1%	0.2%	0.1%	0.1%	0.9%	0.6%	0.9%	0.4%
Paper			2.2%	0.0%	2.1%	0.0%	1.9%	0.0%	2.2%	0.1%
Residual materials			0.1%	0.1%	0.1%	0.1%	0.1%	1.1%	0.4%	0.7%

Obviously, the highest recovered mass of the floating product is recorded for the samples that contained PE bottle bodies and the lower values for the samples that contained sorting faults and residual waste, but a poor correlation was found between the concentration PE of the feedstock and the recovered mass. This is most likely caused by the relative small differences in concentration (89-100%) and the relative large errors in concentration (±2%) and in recovered mass (±2%).

There is a moderate relationship between the recovered mass of the sinking product and the sum of the PS, PVC, PET and other material concentrations in the feedstocks. The moderate correlation is likely to be caused by the small differences in concentration (0-4%) as compared to the error in the concentration (±2%). Although the density separation process effectively removes a sinking product and hence heavy polymers such as PS, PVC and PET, still a substantial part of these heavy polymers is found in the floating product. This is agreement with previous DSC and ATR-IR analysis of comparable PE samples indicating 5-10% PP in the recycled PE [7]. This is also apparent from the good relationship between the concentration of PE in the feedstock and PE in the milled goods, see figure 1. Hence, although this simple standard recycling process is able to remove a part of the contaminants, it is not able to remove them all.

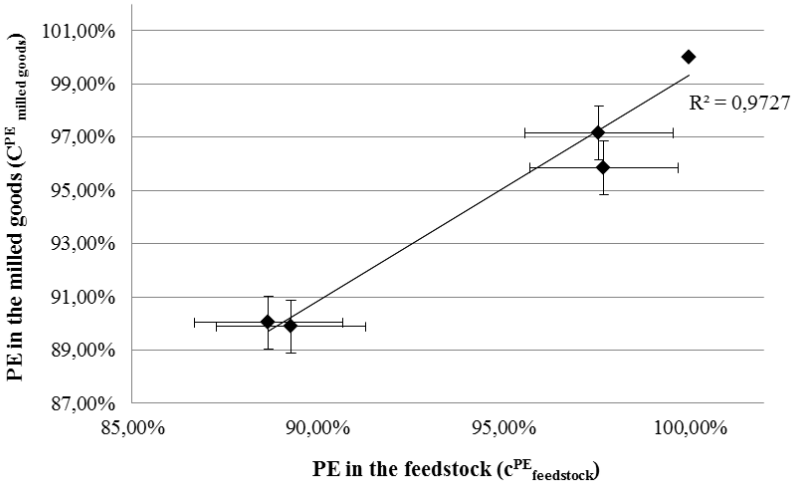

FIGURE 1. The relationship between the concentration of PE in the feedstock and the concentration PE in the milled goods for the six PE samples with a progressively more complex composition.

TABLE (5). Calculated net PE and PO (PE+PP) yields (ηPE and ηPO) for the 6 PE feedstocks.

	1	2	3	4	5	6 warm	6 cold
ηPE	94 ± 3%	101 ± 3%	98 ± 3%	95 ± 3%	97 ±3%	101 ±3%	99 ± 3%
ηPO	94 ± 2%	101 ± 2%	98% ± 2%	97% ± 2%	96% ± 2%	99 ± 2%	98 ± 2%

The net PE yields were calculated with equation 3 and the data in table 3 and table 4 and the net material contents. These net PE yields are listed in table 5. The error in the net PE yields could not be derived with error propagation laws due to the many assumptions made. Instead it was derived from a regression fit of the calculated net PE yields versus the PE concentration of the feedstock (see figure 2), which yielded a residual standard deviation of $\sigma = 2.7\%$ in the net PE yields. Also the net polyolefine yields (PE+PP) were calculated, see table 5. The residual standard deviation was estimated in a similar manner to be $\sigma = 2.4\%$. The net PE yields and the net PO yields are for all samples approximately equal to 100%.

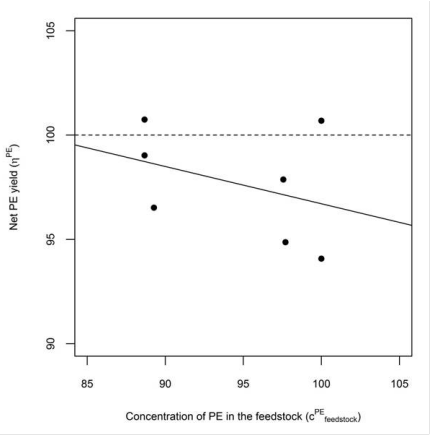

FIGURE 2. The poor relationship between the concentration polyethylene in the feedstock and the net PE yield was used to derive a residual standard deviation in the net PE yield. The dashed line refers to the optimal 100% yield.

A laboratory method for measuring the recovered masses and net polymer yields for the mechanical recycling of sorted plastic products into washed milled goods has been presented. There is a substantial sample-to-sample variation in polymeric composition between similar feedstocks and this variation is also observed in recovered masses. Polymeric contaminants are only partially removed by this standard mechanical recycling process. The net polymer yields are close to 100% for such a standard recycling process. This implies that the recovered mass of recycling can be estimated from a detailed compositional analysis (composition in categories, material composition per category and net material content). Nevertheless, the concentration of polymeric contaminants in recycled plastic still have to be determined by NIR assisted flake sorting.

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