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Circularity Study on PET Bottle-To-Bottle Recycling

Elisabeth Pinter [1], Frank Welle [2], Elisa Mayrhofer [3], Andreas Pechhacker [4], Lukas Motloch [4], Vera Lahme [5], Andy Grant [5] and Manfred Tacker [1,*]

1 Department of Applied Life Sciences, FH Campus Wien, University of Applied Sciences, Helmut-Qualtinger-Gasse 2, 1030 Vienna, Austria; elisabeth.pinter@fh-campuswien.ac.at
2 Fraunhofer Institute for Process Engineering and Packaging, Giggenhauser Straße 35, 85354 Freising, Germany; frank.welle@ivv.fraunhofer.de
3 Department for Microbiology and Cell Culture, OFI–Austrian Research Institute for Chemistry and Technology, Franz-Grill-Straße 5, Object 213, 1030 Vienna, Austria; elisa.mayrhofer@ofi.at
4 Starlinger & Co GmbH, Sonnenuhrgasse 4, 1060 Vienna, Austria; recycling.ph@starlinger.com (A.P.); recycling.mot@starlinger.com (L.M.)
5 Eunomia, 37 Queen Square, Bristol BS1 4QS, UK; vera.lahme@eunomia.co.uk (V.L.); andy.grant@eunomia.co.uk (A.G.)
* Correspondence: manfred.tacker@fh-campuswien.ac.at

Abstract: With the European Green Deal, the importance of recycled products and materials has increased. Specifically, for PET bottles, a high content of recycled material (rPET) is demanded by the industry and consumers. This study was carried out in a lab environment replicating real-life industrial processes, to investigate the possible impacts on rPET quality over eleven recycling loops, aiming to use high amounts of rPET repetitively. A cycle included extrusion, solid state polycondensation (SSP), a second extrusion to simulate bottle production, hot wash and a drying step. 75% rPET and 25% virgin PET were extruded in eleven cycles to simulate a recycling and production process. Samples underwent chemical, physical and biological analysis. The quality of the rPET material was not adversely affected. Parameters such as coloring, intrinsic viscosity, concentration of critical chemicals and presence of mutagenic contaminants could be positively assessed. The quality of the produced material was likely influenced by the input material's high standard. A closed loop PET bottle recycling process using an rPET content of up to 75% was possible when following the proposed process, indicating that this level of recycled content can be maintained indefinitely without compromising quality.

Keywords: circular economy; polyethylene-terephthalate; PET; post-consumer recycling (PCR) material; bottle-to-bottle; recycling; closed-loop

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1. Introduction

The circular economy is of great interest for consumers, legislative bodies and industry alike with the goal of enabling sustainable developments [1,2]. Specifically, plastic packaging has been targeted in the European Union with an emphasis on recycling. Polyethylene terephthalate (PET) is considered to be the most promising food-packaging plastic for recycling and is being used extensively [3,4]. One reason is its ability to absorb post-consumer contaminations at lower levels compared to other plastics, such as polyolefins, therefore making it more suitable for the recycling process [5]. This can be regarded in context with the introduction of super-clean processes, which have been found suitable by the European Food Safety Agency (EFSA) [6–8] to produce post-consumer recycled PET (rPET) safe to be used for food-grade packaging, such as for mineral water.

In 2018, approximately 4.3 million tons of rigid PET packaging were placed on the European market (EU28+2) of which approx. 79% were bottles and the remainder trays. Of this, approximately 1.9 million tons (45%) were collected. In Europe, PET beverage bottles are collected through a combination of deposit return schemes (DRS) and separate

collections. DRS account for approx. 35% of collected PET bottles and the average collection rate for PET bottles in 2018 was 58.2%. Considering losses within the sorting process, an estimated 52% of PET bottles are sent for recycling [9]. Currently, both industry initiatives and regulatory bodies aim to use a higher proportion of recycled PET content to ensure a closed loop system is being maintained. Specifically, bottle-to-bottle recycling for food-grade PET is considered a highly effective way to ensure a high quality of recycled material. Some producers use up to 100% rPET [10] for their bottled products and the use of rPET for plastic packaging in Europe has increased remarkably over the last few years [11]. This being said, the average recycled content in European PET bottles was 11% in 2018 [9].

Several industry initiatives and private sector companies have pledged themselves to increase recycling rates and recycled content in their products [12,13]. There, they target the recycling and reuse of 65% of PET packaging material by 2030, of which 30% is aimed at closed loop [14] and 25% of post-consumer recycling content is the goal for plastics in general by 2025 [11]. Major brands such as Coca-Cola, PepsiCo, Danone and Nestlé have signed up to global commitments set out by Ellen McArthur with Danone and Nestlé exceeding these targets at 50% and 30%, respectively. While some brands are already achieving a 100% recycled content in PET bottles, these bottles, when entering the collection and sorting streams, mix with other bottles which may have much lower recycled content. Hence, the rPET content levels are somewhat diluted. On average in Europe in 2018, bottlers use an estimated rPET content of 11% [9]. The German DRS scheme, Petcycle, which holds approximately 8% [15] of the market share, prescribes to its members the use of at least 75% recycled content (up from 55% in 2020) [16]. The system works mainly in a closed loop in which bottlers receive back their own bottles after consumer use and recycle these with other Petcycle bottles. Considering that closed loop recycling with 55% rPET content is therefore already proven in the market, this study investigates closed loop recycling with 75% rPET content. Furthermore, the EU defined collection targets for beverage bottles at 90% by 2029 [17]. This collection volume would enable a considerably higher rPET content in the bottles at that time. To illustrate an optimistic future scenario, it was decided to perform the study with 75% rPET and 25% virgin PET (vPET).

An important aspect regarding recycling is the presence of contaminants to understand how they might end up in the final product and their consequent possible effect towards consumer health. Specifically carcinogenic, mutagenic or reprotoxic (CMR) substances are of great concern when dealing with unknown substances, such as contaminants, in the context of food contact materials [18]. For this, the International Life Science Institute (ILSI) recommends the combination of chemical analysis combined with in vitro bioassays, specifically with the Ames test [18]. Through this, important additional information can be gained on the toxicological composition of a material. For ensuring consumer safety, several thresholds apply and the exclusion of any risks is an important goal. To produce food grade PET according to EFSA, not more than 5% of feedstock should be derived from non-food grade sources, to ensure a consistent and sufficient quality of products (EFSA 2011). However, a study by Franz and Welle (2020) showed that up to 20% of non-food grade sources does not result in any negative impact or safety concerns [19,20]. By use of a DRS collection system, some of these obstacles can be overcome, as it is ensured that only food contact materials are used in the recycling process. However, such a system does not protect from consumer misuse, which might lead to contamination with non-food applications that could be transferred to the recycling material. In any event, it is important to closely monitor the input recycling material, as contaminations can be more rigorous than for virgin material and can happen at any stage of the recycling chain: at the household use, at collection and sorting, during the recycling process or at the formation of new products [2,21].

Regarding contaminations during the recycling process and renewed production of PET bottles, attention must be given to certain substances. Previously mentioned CMR substances are of utmost concern, but common post-consumer PET substances such as acetaldehyde, benzene, 2-methyl-1,3-dioxolane, ethylene glycol and limonene [22] also

have to be monitored closely and their relevance is explained in the following. Acetaldehyde is a degradation product in PET production, but also a volatile flavor substance found in some beverages. It can be detected by consumers at very low concentrations and often acetaldehyde scavengers or blocking agents need to be added to minimize its amounts [23,24]. Benzene is known to be a reaction product of polyvinyl chloride (PVC) and therefore used to monitor any presence of PVC contaminants [25]. The level of 2-methyl-1,3-dioxolane present depends greatly on the acetaldehyde and ethylene glycol concentration, as it is a condensation product of these substances. The application of solid state polycondensation (SSP) is known to lower the amount of acetaldehyde and ethylene glycol, therefore also minimizing the concentration of 2-methyl-1,3-dioxolane [25]. Ethylene glycol is both a PET monomer and a degradation product [26]. Its concentration depends greatly on the SSP treatment and the process parameters during injection or blow molding [25]. Limonene is considered to be a key indicator for recycling material quality, as it is a typical off-flavor substance and a food-borne contaminant, which levels are greatly reduced in the super-clean PET recycling process [22,27].

When looking at the quality of the input material the presence of contaminating materials or flakes, especially by different oligomers originating from other polymers types, is of great importance. A major aspect when it comes to quality of the rPET material is the sorting and the collection systems providing the input material for recycling. A mono-collection system, where only food-grade PET bottles are collected, was shown to be more efficient and to produce higher quality rPET than co-collection-systems [10]. The same was shown in a study by Alaerts et al. [28], which pointed out that a high quality of the recycling material can be ensured through bottle-to-bottle recycling and that the application of lower grade products, such as soap bottles or textiles, is undesired. In a closed loop approach, the bottle-to-bottle recycling ensures repeated application as beverage food packaging, and the demand for high transparency by the industry can be achieved [29]. The presence of any contaminants leading to hazing or yellowing is undesired. In particular, the presence of other polymers such as PVC or polyamide (PA) contaminants is critical, as these might lead to a yellowing of the recycled PET or to chain degradation, as well as to the formation of benzene [25,30]. Further, any substances affecting the mechanical, physical or chemical quality is undesired, such as contaminations with polyolefins, PET flakes from colored bottles, adhesives, labels or metals, and a high sorting efficiency with pure fractions is the overall aim [31]. However, the presence of impurities or contaminations is not necessarily negative. A study by Welle et al. [32] showed, for example, that the presence of up to 8% PA in PET bottles could positively influence the chemical inertness against sorption. Nevertheless, the yellowing of PET recyclates caused by the presence of PA contaminants [30] can outweigh this as it can lead to an undesired reduction in recyclate quality concerning the coloring of the material.

For PET bottle production, not just optical influences but also intrinsic properties, such as viscosity and melt flow rate of the polymer, are crucial. Specifically, the intrinsic viscosity (IV-value) must be high (>0.8 dL/g) to ensure the polymer chain length is sufficient to allow the bottle formation process [29,30]. The application of state-of-the-art SSP machinery can positively influence the IV value and therefore the processability of the rPET pellets [33]. Through the SSP process, several undesired volatile by-products can be removed and the molecular mass, and therefore indirectly also the IV-value, can be improved [30].

In this study, we aim to simulate a repetitive closed loop system using current industrial processes and to provide insight on the different parameters that might affect the quality of the product. The influence of repetitive product cycles was especially of interest to gain information upon whether the anticipated repetitive use of high levels of rPET in the circular economy would lead to any changes in material quality. The possible contamination through certain process steps was taken into account. Finally, an assessment and interpretation on the quality of the recycled material and the impact of the consecutive recycling cycles was done.

2. Materials and Methods

Characterization of input material: The material used for recycling was obtained from a mono-collection system from Sweden, which is based on a deposit-refund approach and was supplied by Veolia PET (Veolia PET Sevenska AB, Norrköping, SWE). The Swedish DRS covers all beverages made of PET and aluminum including water, soft drinks and certain alcoholic drinks. Only dairy products and specific containers for fruit juices are excluded from the system. In 2019, the total return rate for PET bottles in Sweden was at about 84%, based on the number of bottles on the market [34]. The provided material was automatically and manually sorted, underwent a hot wash process using caustic soda and was extruded by the supplier. Further, the material is a commercially available product and can be considered as representative for the market. To analyze the quality of the input material, a screening method was applied to determine the presence of remaining contaminants. For this, the roast oven test according to DIN EN 15348, annex C was performed, in which 100 g of input material was heated to 210 °C for 30 min under forced ventilation and checked for changes in coloration. This is a visual determination of quality, where the amount and possible presence of contaminants, such as PVC, polyolefins or adhesives is analyzed. Further, the input material was characterized by applying GC-FID analysis with focus on the chemicals: acetaldehyde, benzene and limonene, as described in detail in chemical sample analysis.

Process description: The initial input material for the closed loop system was 100% rPET bottle flakes (Veolia PET Sevenska AB, Norrköping, SWE) from the Swedish DRS sample. In a first step, the PET flakes underwent a recycling process with a recoSTAR PET iV+ (Starlinger, AT), which is used on commercial scale within the industry. This process consisted of a pre-drying step, where the material was pre-dried at 140 °C. Through a 65 mm extruder (~270 °C) with vacuum degassing (<10 mbar), melt filtration (56 µm) and under-water-pelletizing with inline crystallization, new pellets were formed. In the next step, the pellets were treated in the SSP for 7 h (min. 190 °C). Then, the recycled and the virgin fractions were mixed in a ratio of 75% rPET and 25% of vPET, NeoPET 82 (UAB Neo Group, LIT), as can be seen in Figure 1. As described in the introduction, this 3:1 ratio takes an optimistic future scenario into account, where a higher rPET content on the market is assumed. To simulate the bottle production (preforming) the material was processed again through the same extruder and by adding 0.05% of the acetaldehyde blocking agent ATA ADDITIVE 00888 (REPI, S.R.L, IT) which consists of >30%–<40% of the active compound anthranilamide. This blocking agent contains a blue toner to balance the yellowing properties of the blocker. This means, little to no additional blue color additive needed to be used during the process. However, when necessary the colorant ANTI YELLOW REMAP 00090 (REPI, S.R.L, IT) was added to maintain the b* value of the material. Afterwards, the pellets were hot-washed at 75 °C for 15 min with previously used washing water from a commercial washing line for PET flakes in order to simulate the contaminants that would be present if a bottle was placed back onto the market, used by a consumer and collected back through the collection system. The aqueous solution consisted originally of 1% NaOH and 0.2% detergent. The used washing water was removed from an industrial operation (EU post-consumer bottle streams) after more than one week of continuous operation. At the end of each process cycle, the surface moisture from the pellets was removed through drying to its initial weight. The produced material then "re-entered" the recycling process and the steps were repeated. Samples were taken and analyzed after each cycle and in total, eleven cycles were performed. The cut-off of eleven cycles is a pragmatic approach and instead the process could have been performed for an indefinite number of cycles.

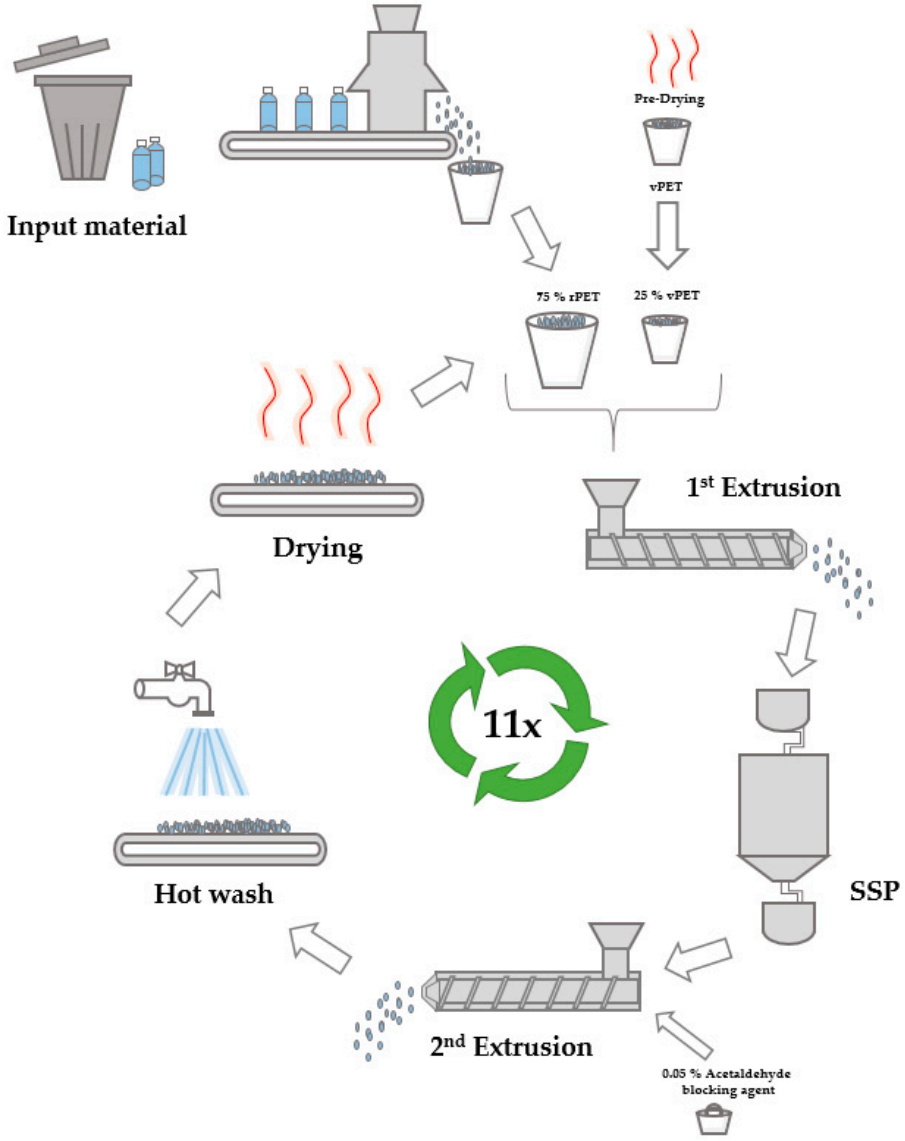

Figure 1. Schematic overview of the simulated recycling process, which consisted of eleven repetitive cycles.

Physical sample analysis: The process was monitored by measuring the process parameters after each cycle through analysis of the pellets and flakes. The color of the pellets was measured through CIE L*a*b* determination using a Ci6X spectrophotometer (X-Rite, Grand Rapids, MI, USA). A sample pool of approximately 10–15 g sample were used and measured in five independent sectors by the device. The calculated mean value derived from the spectrophotometer was obtained, as is industrial standard. Experience values were considered to ensure the process was running smoothly and a suitable endproduct could be obtained. Further, a b* value > 2 was considered to be the cut-off and an anti-yellow agent was added as described above when necessary to maintain the coloring. Moreover, the total color change was measured through calculation of the ΔE value. For this, the following equation was used by comparing the respective L*a*b* values from the cycles with the vPET values: $\Delta E = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$. Further, the melt flow rate (MFR) was measured by using 8 g of sample at 285 °C with 2.16 kg load with a melt flow indexer LMI 5000 (Dynisco, GER) and afterwards, the intrinsic viscosity was converted from the melt viscosity.

Chemical sample analysis: GC-FID analysis was performed for products obtained after each cycle. The samples were analyzed as pellets as delivered without grinding of the pellets. Here the volatile substances acetaldehyde, benzene, limonene, 2-methyl-

1,3-dioxolane and ethylene glycol, were monitored according to the following headspace gas chromatography method: 1.0 g of the samples was introduced in a glass vial and equilibrated at 200 °C for 1 h. After equilibration, the headspace of the vial was injected on a gas chromatography column with a flame ionization detector (FID). Gas chromatograph: Perkin Elmer AutoSystem XL. Column: DB 1–30 m, 0.25 mm i.d. 0.25 µm film thickness. Temperature program: 50 °C (4 min), heat rate 20 °C/min to 320 °C (15 min). The carriage gas was 50 kPa helium and the split was 10 mL/min. Headspace autosampler: Perkin Elmer HS 40 XL. Oven temperature: 200 °C, needle temperature: 210 °C, equilibration time: 1 h, injection time: 0.02 min. The identification was done by retention time and verified with the coupled mass spectrometer. Quantification was achieved by external standards with calibration curves and at least five calibration points per standard. In addition, extraction of the PET materials was performed in order to evaluate non-volatile compounds in the recyclates such as anthranilamide, oligomers and their degradation products. These low volatile substances were analyzed after extraction of the samples with dichloromethane (DCM) as solvent. For each test, 1.0 g of PET material was immersed in 10 mL DCM and stored for 3 days at 40 °C. The extracts were decanted from the PET material and analyzed via gas chromatography with MS and FID. Gas chromatograph: Agilent 6890. Column: ZB 1MS-30 m, 0.25 mm i.d. and 0.25 µm film thickness. Temperature program: 50 °C (2 min), heat rate 10 °C/min to 340 °C (10 min). For quantification of anthranilamide, another column was used: Column: ZB WAXplus-30 m, 0.25 mm i.d. and 0.25 µm film thickness. Temperature program: 50 °C (2 min), heat rate 10 °C/min to 240 °C (14 min). In both cases, the pre-pressure was 50 kPa hydrogen gas and the split 1:20. Two internal standards were used: butyl hydroxyanisole (BHA) and Tinuvin 234. Identification and characterization were achieved by coupling the headspace chromatography to a mass spectrometer.

Biological sample analysis: The input material and samples from cycles 1, 5 and 10 were further analyzed for possible adverse health effects of contaminants. For this, the approach suggested by the ILSI [18] was followed. The samples were extracted with DCM for 72 h at 37 °C, shaking at 150 rpm using a ratio of 30 g of granulate per 300 mL of solvent. A similar protocol was used by Franz and Welle [19] for extraction of non-food PET bottles. For samples with very high particle content, a filtration step using glass fiber filters was applied after the extraction. In a next step, the extract was concentrated through parallel evaporation by a factor of 300 using a Syncore® Analyst device (BÜCHI, GER). Following parameters were applied: platform temperature: 55 °C, cover temperature: 50 °C, rotation: 300 rpm, pressure gradient: 20 min at ambient pressure, 2 min at 800 mbar, 2 min decreasing from 800 to 650 mbar, 1.5 h at 650 mbar. Evaporation was stopped when <5 mL of DCM remained. Dimethyl sulfoxide (DMSO) was added as a keeper and the remaining DCM was then removed by evaporation decreasing the pressure down to a minimum of 50 mbar until only the sample dissolved in DMSO remained. For each sample, two independent sample extracts were prepared. Furthermore, a solvent blank was prepared, treated equally to the samples throughout the whole process. The concentrated samples were then applied in the Ames MPF™ (Xenometrix, Aschwili, CH) following the protocol suggested by Rainer et al. [35]. For S9 experiments, phenobarbital/β-naphthoflavone induced S9 was used provided by Xenometrix (CH). Spiking experiments with known mutagens were performed to gain information on possible interactions of the sample matrix with the bacterial test system and the presence of growth inhibiting and/or bacteriotoxic substances.

Bottle production: The material produced after cycles 1, 4, 7 and 10 was used for bottle production. This was done to determine whether the produced material was not just theoretically, but also practically suitable for bottle production. For this, 10 kg of sample were provided. Using a single stage injection/blow molding machine Automa NSB50 (Preven, IT), 500 mL bottles were produced with a weight of 23 g and a two-cavity mold was used to produce the preform of the type PCO 1810. At the single stage machine, the production took place in two steps: first, the PCOs were produced and then the blow molding was done to produce the final bottles. The following parameters were used: 19.3 s cycle time, injection time of 0.98 s, and cooling time from 2.2 to 2.4 s. The temperature in

the extruder ranged from 276 to 285 °C and the mold temperature was 285 °C. The injection pressure was 50 bar, with a hold pressure at 45 bar. During blowing, a compression time of 2.5 s at a pressure from 10 to 40 bar for 1.4 s was used.

3. Results

3.1. Intrinsic Viscosity and Optical Quality

In the roast oven test, the input material showed no presence of PVC, polyolefins, polyamides, metals and multilayered or PET flakes from colored bottles. The analysis of 100 g sample detected a glue content between 990 to 2740 ppm. This corresponds to 0.1 to 0.27% of glue in the 100 g sample, which is considered to be well below any industry best practice threshold. Further, minor quantities of labels and yellow PET flakes were found, which were considered neglectable. To sum up, the results of the roast oven test showed a very high quality and purity of the input material, as was expected from a mono-collection deposit system.

For color determination, the CIE L*a*b* system was used. All samples were measured after the SSP step. Especially important for rPET recycling is the graying, which can be measured with the L* value, and the yellowing, measured by the b* value. A positive b* value is associated with undesired yellowing, and a high L* value means little graying of the polymer flakes [36]. For the b* value, a cut-off of >2 was used to determine whether too much yellowing took place. The results of this analysis are shown in Table 1. The b* value was well controlled through the addition of an anti-yellow agent and the blue coloring of the acetaldehyde-blocking agent, when necessary. During the applied process the b* value ranged from 0.14 to 1.34. The same is true for the L* value, which was very stable throughout the circularity study with values from 66.23 to 71.02, indicating hardly any graying taking place.

Table 1. Overview of the results for the physical characterization of the samples. Testing was done after SSP for each cycle regarding MFR, IV and color values.

Sample Code	IV [dL/g]	CIE L*a*b* Color Value			
		L*	a*	b*	ΔE
vPET	0.820	84.44	−2.02	−1.51	
rPET Flakes	0.778	-	-	-	
Cycle 1	0.870	69.76	−2.59	0.92	14.89
Cycle 2	0.854	69.74	−2.80	0.47	14.85
Cycle 3	0.830	66.23	−2.73	0.14	18.30
Cycle 4	0.845	69.24	−2.87	0.59	15.37
Cycle 5	0.835	68.67	−2.86	1.35	16.05
Cycle 6	0.804	69.55	−2.82	1.05	15.13
Cycle 7	0.859	69.72	−2.90	0.40	14.87
Cycle 8	0.801	69.88	−3.00	0.59	14.74
Cycle 9	0.781	70.06	−2.75	0.76	14.58
Cycle 10	0.797	68.53	−2.70	0.54	16.06
Cycle 11	0.809	71.02	−2.68	0.30	13.56

Furthermore, the IV values were monitored, as they have great importance for processing. A high IV value (>0.8 dL/g) is required for bottle production. For carbonated drinks, even higher IV values are desired [29], which can be up to 0.84 dL/g. Table 1 shows the IV values of the input material, the vPET and the rPET after each cycle. The IV value of the input material (0.778 dL/g) is slightly below the required 0.8 dL/g. The IVs in the process are rather high and even after several cycles the criteria can still be met. However, after nine cycles there is a slight decrease in IV, which could be compensated through alterations of process parameters within the SSP process. In continuous production, it would be easier to adjust the parameters according to the requirements. During the study, the processing time for the recycling step was less than 1 h and therefore limited the adjustments.

3.2. Chemical Quality

Chemical analysis was conducted to determine the presence of undesired substances. These could possibly originate from side reactions or contaminations, e.g., with wash water or other process steps, and could accumulate during the whole recycling process. In this study, we focused on the substances: benzene, AA, limonene, ethylene glycol and 2-methyl-1,3-dioxolane. The quantified results are shown in Table 2. For two of these substances, migration thresholds can be derived from the European regulation no 10/2011; these are 6 mg/kg for AA and 30 mg/kg for ethylene glycol [37]. However, it has to be noted that these thresholds apply to migration studies into food simulants. In general, the substances are below any best practice thresholds or limits used as quality parameters. For rPET intended for food contact, the EFSA suggests a contamination level of 3 mg/kg in washed flakes before super-clean recycling [38] for post-consumer substances as well as for substances from misuse of the PET bottles for household and garden chemicals. The concentrations determined in this study are below this concentration in any cycle of this study.

Table 2. Results of the chemical analysis of the input material and after the cycles 1 to 11. The volatile substances were detected with a targeted analysis through GC-FID.

Sample	Acetaldehyde [mg/kg]	2-Methyl-1,3-dioxolane [mg/kg]	Benzene [mg/kg]	Ethylene Glycol [mg/kg]	Limonene [mg/kg]
rPET flakes	8.2 ± 4.71	2.8 ± 1.0	0.25 ± 0.25	6.7 ± 1.5	3.2 ± 1.0
Cycle 1	1.0 ± 0.1	0.12 ± 0.1	<0.03	0.77 ± 0.02	<0.03
Cycle 2	1.2 ± 0.1	0.12 ± 0.1	<0.03	0.66 ± 0.03	<0.03
Cycle 3	1.5 ± 0.1	0.14 ± 0.1	<0.03	0.70 ± 0.01	<0.03
Cycle 4	1.3 ± 0.1	0.13 ± 0.1	<0.03	0.61 ± 0.01	<0.03
Cycle 5	1.7 ± 0.1	0.14 ± 0.1	<0.03	0.88 ± 0.01	<0.03
Cycle 6	1.7 ± 0.1	0.16 ± 0.1	<0.03	0.59 ± 0.04	<0.03
Cycle 7	1.4 ± 0.1	0.14 ± 0.1	<0.03	0.54 ± 0.01	<0.03
Cycle 8	2.1 ± 0.1	0.14 ± 0.1	<0.03	1.12 ± 0.01	<0.03
Cycle 9	1.6 ± 0.1	0.13 ± 0.1	<0.03	0.92 ± 0.04	<0.03
Cycle 10	1.4 ± 0.1	0.12 ± 0.1	<0.03	0.75 ± 0.01	<0.03
Cycle 11	1.4 ± 0.1	0.12 ± 0.1	<0.03	0.73 ± 0.03	<0.03

For the input material, the results were obtained in two independent methods (a) headspace gas chromatography (GC) for analysis of volatile substances and (b) extraction of the recyclates followed by gas chromatographic analysis of the extracts. The chemical analysis was performed on three subsamples, where each time 1.0 g of sample was measured. As a result, for the volatile substances determined by headspace GC compared to other studies, the concentration of acetaldehyde, benzene, 2-methyl-1,3-dioxolane, ethylene glycol and limonene is well within the average [19,25]. Further, a relatively high deviation can be seen in the input material (rPET flakes) in Table 2, which is quite typical for post-consumer PET flakes. The pellet samples are much more homogeneous and show significantly lower standard deviations in the three subsamples. However, the concentrations found in the input materials are low, which indicates that the input material can be classified as very high quality. All substances are quantified in lower concentrations in the recycled samples even after eleven recycling cycles.

Results of the extraction on anthranilamide and oligomers showed that the highest concentrations of the substances were found in the input material (rPET flakes, Table 3). The pre-dominant substance in the samples was the PET cyclic trimer. In agreement with the results for the high volatile substances, also the low volatile substances are determined in lower concentrations as in the input material. After each cycle, the acetaldehyde-blocking agent anthranilamide was added with a concentration of 500 mg/kg. Anthranilamide reacts with acetaldehyde to form 2-methyl-4-hydroxy-1,3-diazanaphthalin (2-methyl-4-hydroxy-chinazolin) [39]. Typical concentrations of anthranilamide in the PET bottle wall

were found between 190 and 320 mg/kg [40]. This shows that the applied approach is quite realistic for the application of mineral water bottles. As a result, both anthranilamide and 2-methyl-4-hydroxy-1,3-diazanaphthalin were efficiently decontaminated during the recycling steps, which is due to the high cleaning efficiency of the applied super-cleaning recycling process. Moreover, the PET oligomers are significantly reduced in concentration during the eleven recycling cycles. PET oligomers are by-products formed during the PET polymerization process. The results of this study show that during recycling the oligomers are not re-generated.

Table 3. Results of the chemical analysis of the input material and after the cycles 1 to 11. The low volatile substances were detected with a targeted analysis through GC-FID after extraction with DCM.

Sample	Anthranil Amide [mg/kg]	PET Dimer [mg/kg]	PET Dimer [mg/kg]	PET Dimer [mg/kg]	PET Substance [mg/kg]	PET Trimer [mg/kg]	PET Trimer [mg/kg]	PET Trimer [mg/kg]
rPET flakes	10.0 ± 0.4	51.9 ± 4.7	26.8 ± 0.7	130.7 ± 7.5	11.4 ± 0.5	119.7 ± 9.2	4310 ± 138	81.6 ± 11.9
Cycle 1	<5	15.5 ± 1.3	10.5 ± 0.5	53.4 ± 2.6	4.0 ± 0.7	40.6 ± 1.9	1370 ± 58	20.9 ± 0.9
Cycle 2	<5	16.5 ± 2.3	11.3 ± 0.6	56.0 ± 0.4	3.9 ± 0.2	43.7 ± 0.8	1490 ± 12	24.1 ± 2.2
Cycle 3	<5	15.2 ± 1.2	9.0 ± 0.4	47.7 ± 0.7	3.2 ± 0.1	38.2 ± 0.9	1340 ± 25	19.6 ± 2.0
Cycle 4	<5	16.2 ± 3.6	11.1 ± 0.5	57.9 ± 2.3	4.0 ± 0.2	45.4 ± 1.8	1600 ± 17	24.3 ± 1.4
Cycle 5	<5	19.6 ± 1.3	12.5 ± 0.7	66.2 ± 3.2	4.2 ± 0.5	50.0 ± 0.7	1800 ± 38	29.2 ± 1.8
Cycle 6	<5	17.7 ± 1.9	11.0 ± 0.5	59.3 ± 3.3	3.7 ± 0.2	44.5 ± 1.6	1620 ± 55	26.1 ± 1.0
Cycle 7	<5	16.8 ± 1.5	10.8 ± 0.3	59.5 ± 1.3	4.1 ± 0.5	44.4 ± 0.5	1650 ± 15	27.3 ± 2.2
Cycle 8	<5	16.1 ± 2.3	11.2 ± 0.6	61.7 ± 2.9	4.0 ± 0.2	46.2 ± 1.1	1710 ± 35	28.1 ± 1.7
Cycle 9	<5	16.5 ± 1.4	10.4 ± 0.7	55.3 ± 0.4	3.6 ± 0.4	41.9 ± 1.1	1570 ± 38	24.1 ± 2.5
Cycle 10	<5	14.9 ± 2.3	9.8 ± 0.5	52.5 ± 2.2	3.8 ± 0.2	40.2 ± 1.0	1520 ± 34	25.1 ± 3.0
Cycle 11	<5	13.2 ± 1.8	8.0 ± 0.7	42.1 ± 2.2	2.9 ± 0.2	34.2 ± 1.3	1280 ± 36	21.4 ± 2.7

Further, the results show that there is no accumulation or formation of the described key substances during the process. Through the process temperatures and the application of the SSP, these substances tend to evaporate, as they are volatile. Moreover, the precise control of the process temperatures ensures that there is no increase in any of these contaminants through degradation processes. In a nutshell, any of these substances present in higher concentrations in the input materials could be lowered during the process, therefore showing its efficiency in producing high-quality recycling material.

3.3. Biological Quality

The input material and the samples taken after cycle 1, 5 and 10 were used for analysis with the Ames MPF^{TM}. In order to allow bioassay testing, concentrated DMSO extracts were produced from these materials. Mutagenicity testing was performed using the *Salmonella typhimurium* strains TA98 and TA100 with and without metabolic activation by S9 rat liver extract. Through the application of the Ames MPF^{TM}, possible mutagenic effects caused due to the presence of direct DNA-reactive genotoxic substances should be detected. The results in Figure 2A–D show that no genotoxic substance could be determined, neither in the input material nor in the cycle samples, as the response was below the threshold for mutagenicity as shown for two independent extracts per sample.

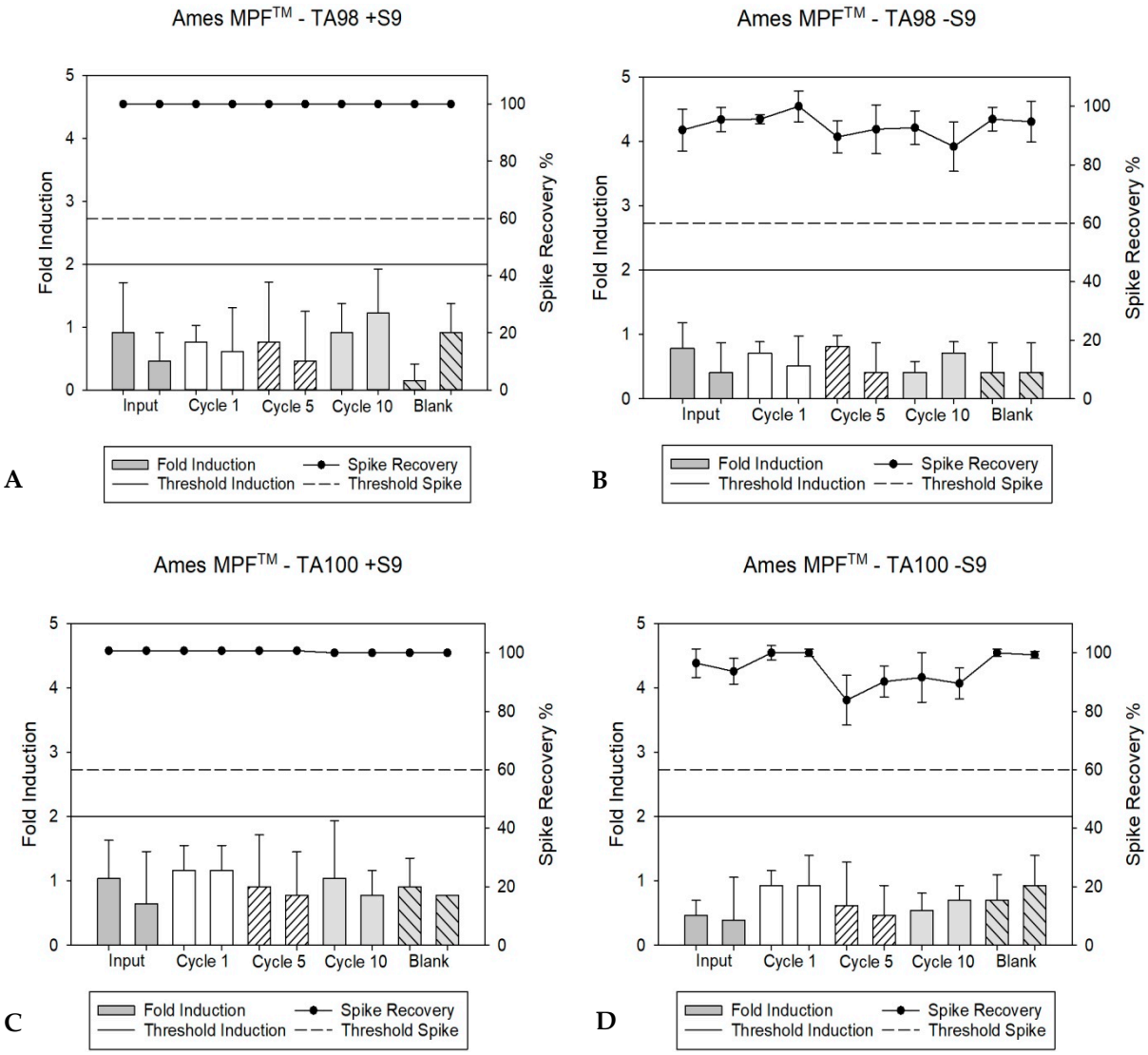

Figure 2. Results of the Ames MPF™ for the different samples. Experiments were conducted with the strains TA98 with (**A**) and without S9 (**B**) and TA100 with (**C**) and without S9 (**D**). The left y-axis represents the fold induction, where the sample mean is compared to the mean background signal and the respective increase is shown. A threshold of two is used as a cut-off for positive results. The right y-axis represents the recovery of a known spike concentration with known mutagens. A threshold of 60% signal recovery compared to the spiked control was used to determine any inhibiting effects. Two independent extracts of each sample were analyzed represented by one bar each.

Moreover, spiking experiments with known genotoxic substances were done to ensure that the presence of the sample matrix did not lead to any inhibiting effects which could conceal a mutagenic response. As presented in Figure 2A–D, the samples showed sufficient recovery when spiked with mutagenic positive controls, indicating that the assay performance was not negatively influenced by the samples.

3.4. Bottle Production

The results of the bottle production through injection/blow molding are shown in Figure 3. The produced sample material was used for bottle production, to show that the extrusion process is possible with the obtained material. The results display that the material had the required characteristics for the production of 500 mL beverage bottles through injection/blow molding.

Figure 3. Produced 500 mL PET bottles from cycle 1, 4, 7 and 10 (right to left) through an injection blow molding process.

4. Discussion

With the European Green Deal published in 2019 [41], the movement of the EU towards a circular economy has been manifested. A literature study by Kirchherr et al. (2017) [1] showed that no uniform definition of a circular economy exists and that several definitions only concentrate on recycling. Simply focusing on recycling is not the sole purpose of the circular economy, but their findings show the importance of recycling in the scheme and broader sense. As such, the concept of the circular economy and sustainability recycling and recyclability have to be considered and are a key factor to ensure a circular approach [42].

Specifically PET recycling and the use of rPET for beverage bottles has been well-established in the last several years [3]. Therefore, it is assumed that the requirements to increase the recycling content for all packaging material on the European market will be possible in the PET bottle sector, through the success of advanced collection and sorting systems and the bottle-to-bottle recycling. Nevertheless, the question arises of how multiple recycling loops will affect the quality of the PET material and bottles produced. So far, simulation studies have been done on the topic of repetitive recycling loops, which concluded that the control of critical parameters and a high quality of input material is of uttermost importance [10]. Discoloration or a change in color is considered a problematic parameter for mechanical recycling of PET [36,43]. In the recycling process, this can lead to the undesired yellowing or hazing of rPET material. Therefore, the L* and b* parameters have to be closely monitored. To prevent yellowing, a good input material is essential, with little contamination by polymers causing yellowing, such as PA. Moreover, a blue colorant can be added to counteract the yellowing process. In this study the b* value, characteristic for blue or yellow color, could be well controlled and hazing was not an issue. Therefore, the process continuously resulted in transparent flakes with good optical properties, which is an important criterion for bottle manufacturers. Another important characteristic for PET recycling is the IV-value. As already stated, for bottle production an IV of >0.8 dL/g is considered to be sufficient [29,30]. The results of this study clearly showed that high IV values were possible even after several cycles. Further, through bottle production the technical applicability could be shown. During the bottle production, residence times were considerably longer than typically applied in practice in industrial processes [44]. This can be considered as a worst-case scenario since longer residence times or repeated thermal

processing [30] lead to a higher thermal load, possibly resulting in yellowing, which was not observed with the produced material.

Volatile substances are especially critical, as they might migrate into the packaged foodstuff. The obtained results for the chemicals acetaldehyde, ethylene glycol and 2-methyl-1,3-dioxolane are similar to those found in other studies on rPET, where a high quality of the material was concluded [25,29]. Acetaldehyde is a common off-odor substance identified in mineral water and therefore blocking agents are added to minimize the formation of acetaldehyde [23,45,46]. Moreover, limonene, which is considered as a critical sensory parameter, and benzene, a typical contamination parameter [25], were found well below any critical concentrations. Further, the screening for low volatile contaminants showed no critical concentrations of any detected substances. The detected concentrations are significantly lower than in the input materials. The acetaldehyde blocking agent anthranilamide and its reaction product 2-methyl-4-hydroxy-1,3-diazanaphthalin were efficiently removed during recycling. Even though it was added in each of the eleven cycles, the additive does not accumulate with increasing recycling cycles.

The substances or possible contaminants monitored through chemical analysis in this study are considered not to be carcinogenic, mutagenic or reprotoxic (CMR) and are therefore unlikely to pose a risk towards consumer safety [47]. To obtain more information on the overall effect of the substance mixture, the ILSI recommends performing additional testing with in vitro bioassays [18]. Specifically the Ames test is suggested to detect any mutagenic effects originating from the material and has shown to be the most suitable method in recent studies [35,48,49]. In this study the material samples were concentrated and applied in the Ames MPF^{TM}, as has been successfully done in a previous study by Rainer et al. [35]. The results clearly showed that no positive response concerning mutagenicity was obtained. Further, any effects caused by the sample matrix towards the bacteria did not lead to inhibiting effects, and therefore it can be concluded that no mutagenic effect was masked through this. Even though the Ames test has superior performance compared to other genotoxicity bioassays regarding detection limits, the lack of sufficient analytical sensitivity of this test [18,48,49] has to be noticed and the obtained negative results have to be regarded with caution.

Finally, the quality of the input material is considered to greatly affect the recycling process and the consequent application of the post-consumer recycling material [10,29,50]. It has been concluded by McDonough and Braungart [51] (p 56) that "most recycling is actually downcycling [as] it reduces the quality over time". This assumption has been closely monitored in this study, and the aim to preserve a high quality and prevent downcycling was of uttermost importance. Quality preservation was analyzed through comparison of the material properties over the consecutive cycles. The IV value and (dis-)coloration was also considered and compared to requirements for virgin material to ensure they were in permissible ranges. Further, the use of input material derived from a mono-collection system with a DRS approach ensured a high quality of the input material. In this study, it was shown that the quality of the rPET material was comparable to the mechanical quality of a virgin PET material. Even after eleven cycles, no loss of mechanical quality could be observed and the presence or accumulation of the monitored substances was satisfactory. Further, any changes in quality could be controlled with industry best practice control measures, e.g., through changes of SSP parameters or addition of colorants. This leads to the assumption that the recycling material produced through state-of-the art commercially used processes has a consistent quality. However, no estimations can be made, whether this is also the case for input material with a different (lesser or even better) quality and how changes in the process might affect this. Moreover, the maximum cycle number should not be seen as a threshold for recycling quality. For this specific process and input material, more cycles might have been possible.

5. Conclusions

The application of post-consumer recycling content in food packaging material is an important contribution to the circular economy. Bottle-to-bottle PET recycling is a prime example for the application of rPET and an interesting solution for the industry, if quality parameters can be met. This study demonstrated that even after eleven cycles the quality criteria of rPET suitable for bottle production could be achieved.

The results of this study underline the potential of rPET to fulfill all tested parameters for a re-use after multiple loops. The trends of the results within the study would not give any indications about a limitation of the number of cycles at the tested ratio of 75% rPET and 25% vPET. In case of a collection rate of 90% of PET bottles in 2029, this ratio could potentially result in a re-use of all produced rPET in the same market. Nevertheless, the authors assume that the outcome is influenced by the purity of the input material, which must be of high and relatively consistent quality, as well as by the process parameters and characteristics, which were based on real life commercial operations.

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