

# Effect of recycled content and rPET quality on the properties of PET bottles, part I: Optical and mechanical properties

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The effect of recycled content and type of recycled poly (ethylene terephthalate) (rPET) on the haziness; colour parameters  $L^*$ ,  $a^*$ , and  $b^*$ ; total colour change; ultraviolet-visual spectrum (UV-VIS) absorption; and environmental stress cracking (ESC) of PET bottles was studied. Three series of PET bottles were made at a small scale production facility with same type of virgin PET and three different types of rPET with recycled contents of 25%, 50%, 75%, and 100%. Also, the particle contamination of the produced PET bottles was studied by dissolving bottle fragments and counting the insoluble particles. A linear relationship was found between the haziness of PET bottles and the particle contamination, with a coefficient of determination of 0.96. Linear relationships were also found between the colour parameters  $L^*$  and  $b^*$  and the particle contamination of the PET bottles, but the slope differed for each type of rPET. The origin of rPET (mono-collection or cocollection) seems to be the crucial factor determining particle contamination and colour of the produced bottles. No correlation was found between the recycled content and the incidence of ESC. However, PET bottles in which high intrinsic viscosity (IV) values were measured showed lower chances of ESC than PET bottles with low IV values

## KEYWORDS

colour, environmental stress cracking, haze, particle contamination, PET bottles, recycling

## 1 | INTRODUCTION

Poly (ethylene terephthalate) (PET) is a widely used packaging polymer for bottles. Most PET bottles are used for water and beverages, but other uses include oils, vinegar, detergents, and household chemicals. The popularity of PET bottles stems from their optical transparency, moderate water vapour transmission rates, and low oxygen and carbon dioxide gas transmission rates. In recent years, the use of recycled PET (rPET) in bottles has increased largely in Europe<sup>1</sup> as the solid state

postcondensation (SSP) became available to ensure the food safety of the rPET material,<sup>2</sup> and the environmental benefits of using rPET instead of virgin PET for bottles<sup>3,4</sup> became increasingly accepted. With the increased usage of rPET in bottles, several technical downsides also became more apparent, such as the reduced optical transparency and their more grey and/or yellow colours.<sup>5</sup> The term haze is used to refer to the reduced optical transparency of rPET,<sup>6</sup> and the colours of rPET are often measured with CIELAB method.<sup>7</sup> Besides the optical downsides of rPET, some beverage producing companies claim that

**Nomenclature:** ABS, Acrylonitrile butadiene styrene; ESC, Environmental stress cracking; EVOH, Ethylene vinyl alcohol; GC, Gas chromatography; GPC, Gel permeation chromatography; HFIP, Hexafluoro-iso-propanol; IR, Infrared; IV, Intrinsic viscosity; NIAS, Nonintentionally added substance; NMR, Nuclear magnetic resonance; PE, Polyethylene; PET, Poly (ethylene terephthalate); PP, Polypropylene; PPTI, Particles per 10 000 images; PS, Polystyrene; PVC, Poly (vinyl chloride); RC, Recycled content; rPET, Recycled poly (ethylene terephthalate); SBM, Stretch blow moulding machine; SSP, Solid state polycondensation; UV-VIS, Ultraviolet-visual spectrum

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the incidence of mechanical bottle failure, as evaluated by environmental stress cracking (ESC), is higher for PET bottles with recycled content. However, this has not been reported in literature. In contrast, the mechanical properties of PET sheets have been reported to be insensitive to the recycled content levels<sup>8</sup> although they were highly degraded when exposed to chemical reagents.<sup>9</sup>

The above mentioned optical and mechanical properties together with migration of nonintentionally added substances (NIAS) might limit the application of recycled content in PET bottles. This raises the question whether these critical properties of PET bottles pose limits on the use of recycled content. Migration of NIAS from PET bottles with recycled content to water is studied in a separate paper.<sup>10</sup>

Haze is caused by the internal randomly scattering of light, which is determined by crystallinity, optical defects, blend formation, and the dispersion of particles.<sup>6</sup> PET bottle walls contain very small highly aligned crystals produced by strain induced crystallisation resulting in highly transparent bottles.<sup>11</sup> Sometimes, larger crystals are formed due to uncontrolled crystallisation in bottom and neck parts of the bottles resulting in localised haze. In most cases, the processing can be adapted to avoid localised haze. Therefore, haze in PET bottle walls with recycled content is most likely caused by the present particles. The particles normally found in mechanically rPET originate from foreign materials (paper, metals, glass, organics, environmental dust, and sand), from additives, and from different immiscible polymers (polystyrene [PS], poly (vinyl chloride) [PVC], polyethylene [PE], polypropylene [PP], acrylonitrile butadiene styrene [ABS], etc) that were incompletely removed during the sorting and recycling process steps.<sup>12-16</sup>

The grey colour of rPET is most likely caused by the presence of particle contamination and can be further worsened by the deliberate addition of reheat additives and blue colorants to mask yellowing.<sup>13</sup> Greying is quantified with CIELAB colour measurements as a reduction in the  $L^*$  colour parameter or alternatively as a relative increase in the thickness corrected ultraviolet-visible spectrum (UV-VIS) absorption values over all wavelengths.<sup>8</sup>

rPET tends to have a yellow colour due to degradation reactions in the polymer backbone. Two main mechanisms have been proposed: oxidation of the diethylene glycol comonomer resulting in hydroquinone and quinone moieties and  $\beta$ -scission of the ester moieties.<sup>17-24</sup> Besides quinones, also other conjugated structures can be formed during degradation reactions such as benzophenones, stilbenes, and biphenyls within the polymer backbone and these can also colour the rPET more yellow.<sup>23,24</sup>

Repetitive heating of PET samples has been reported to cause a reduction in the mechanical properties.<sup>25,26</sup> This is most likely caused by chain scission reactions and subsequent morphological changes. In rPET treated with SSP, the chain lengths are restored, and hence, a loss of mechanical properties is less likely. However, several polymeric contaminants (PVC, PS, EVOH, and PP) in rPET retard the restoration of the chain lengths during this treatment.<sup>13</sup> A highly undesired type of mechanical failure of PET bottles is ESC in which residual stresses in the bottom part of the bottle are suddenly released by the formation of a puncture or a crack, after which the bottle loses its contents. This effect is mostly found in bottles

with a petaloid base.<sup>27</sup> Although substantial research efforts have been undertaken, the causes of ESC are still not completely understood.<sup>28</sup> ESC is more often witnessed at higher levels of humidity and at higher levels of alkalinity. This suggests that the underlying mechanism involves ester bond hydrolysis in the stressed parts of the PET bottle.<sup>29</sup> Furthermore, PET bottles made from resins with lower intrinsic viscosities (hence lower molecular weights) were also found to experience more ESC.<sup>30</sup> However, no scientific publication relates ESC to the use of rPET, yet

This study aims to improve our understanding of critical bottle properties, underlying mechanisms and influencing material parameters with the intention to find correlations between these properties and parameters. This is approached by studying a matrix of PET bottles with various levels of recycled content and various qualities of rPET from two different collection systems and systematically studying these with regards to particle contamination, IV and the critical optical properties haze, colour and UV-VIS absorption, and the mechanical property ESC.

## 2 | MATERIALS AND METHODS

### 2.1 | Materials

Three types of rPET pellet and one type of virgin PET resin materials were used to produce PET bottles. rPET pellet materials were received from beverage companies. They were screened on colour ( $L^*$ ,  $a^*$ , and  $b^*$ ) and volatile substances (acetaldehyde, benzene, limonene, and toluene) by the gas chromatography (GC) headspace measurements (ASTM F2013-10). According to these results, the received rPET materials were named A, B, and C. Type A rPET was primarily made from mono-collected PET bottles, its benzene concentration was below the detection limit (0.05 mg/kg), the acetaldehyde concentration was 0.39 mg/kg and its  $b^*$  colour was  $-4.6 \pm 0.2$ . Types B and C rPET were primarily made from cocollected PET bottles. Pellets of type B had an acetaldehyde concentration of 1.08 mg/kg, a benzene concentration of 0.05 mg/g, and a  $b^*$  value of  $2.2 \pm 0.3$ . Pellets of type C had an acetaldehyde concentration of 3.68 mg/kg, a benzene concentration of 0.44 mg/kg, and a  $b^*$  value of  $-1.5 \pm 0.3$ . Because of the high amount of acetaldehyde present, type C will not be used by beverage companies. Virgin PET RAMAPET N180 was supplied by Indorama. An outline of its specification is listed in Table SA3 (Supporting Information). The acetaldehyde and benzene concentrations in the virgin PET pellets were 0.79 and 0.08 mg/kg, respectively. The  $b^*$  value of the virgin pellets was  $-4.2 \pm 0.2$ .

### 2.2 | Production of bottles

The three types of rPET pellets were mixed with virgin PET pellets in weight ratios of 25:75%, 50:50%, and 75:25% in amounts of 7.5 kg. Materials were pre-dried at 160°C for 6 hours and injection moulded to produce preforms in a DEMAG 50 Ton machine at 555 bar injection pressure with an average cycle time of 33 seconds.

The extrusion barrel had five temperature zones, in sequence from barrel to mould: 40°C at the hopper, 270°C, 280°C, 290°C, and 285°C at the die. The temperature in the mould was kept constant at 25°C. The preforms had an average weight of 19 g. Roughly, 100 preforms were produced for each mixture of virgin PET and rPET. Also, preforms of 100% virgin material and 100% rPET types A, B, and C were made. All preforms were produced under identical conditions. A technical drawing of the preform can be found in Figure SA1 (Supporting Information).

Bottles were blown from the preforms in a set-up consisting of a Shenzhen J&D preform reheating unit and a Shenzhen JND-880 semi-automatic stretch blow moulding machine (SBM) provided with a steel mould cavity. The preform was heated in 165 seconds to an even temperature of around 110°C above the neck. In the reheater, the electrical current to six different infrared (IR) lamps had to be adjusted according to the material been produced; the darker colour of the preforms containing RC influences the absorption of IR radiation.<sup>5</sup> Applied voltages are reported in Table SA1 (Supporting Information). The bottles were produced in two separate runs using the same preforms. In the first run, preform temperatures could not be monitored; however, in the second run with the same machine settings, a thermal imaging IR camera was used to monitor the actual temperature of the preform before transferring it to the SBM. Measured temperatures are reported in Table 4. Bottles produced in the second production run were used mainly for ESC tests. The heated preforms were manually transferred to the SBM in about 20-second time. The conditions of the SBM were kept constant for all different materials and mixing ratios: 26 bar of air pressure, 12 s cycle time, and a mould temperature of 20°C. A technical drawing of the bottle shape can be found in Figure SA2 (Supporting Information). Data on the wall thickness and material distribution of the produced bottles are collected in Table SA2 (Supporting Information).

## 2.3 | Characterisation of bottles

### 2.3.1 | Particles in solution

Particles in solution (partisol) measurements were performed at Emmtec with the Micro Scout system of Eagle Vision Systems BV. This image analysis method was developed to detect solid (insoluble) micro particle contaminations in PET materials used for fibre spinning.<sup>16</sup> Nowadays, it is used to evaluate the quality of raw PET materials used in different production processes. One bottle per type was analysed. The bottom of the bottle was removed, strips were taken from the bottle walls, and cut into small pieces of around 1 × 1 cm. These square pieces were washed with 0.2 µm filtered ethanol to remove external dirt. Then, they were blown dry and dissolved in hexafluor-iso-propanol (HFIP) at a concentration of 10 g/100 mL. This solution was pumped through a cuvette; 10 000 images were taken and analysed. Counted particles were classified in five groups according to their size in µm: 0 to 3, 3 to 5, 5 to 10, 10 to 25, and 25 to 100. Since most particles are not perfect spheres, this size distribution is only indicative. Also, specific types such as transparent particles,

thread like types and gel like types were identified separately and classified by size. A sensitivity value of 45 was used as pixel threshold for all measurements in bottles. Here, the total amount of measured particles per 10 000 images (PPTI) is reported, since it is a single measurement, no standard deviations are available.

### 2.3.2 | Intrinsic viscosity

The intrinsic viscosity (IV) was determined according to ASTM D4603-03. 250 mg PET bottle wall pieces were dissolved in 50 mL phenol/tetrachloroethane (60/40 w/w) at 100°C for 30 minutes. The viscosities were determined in calibrated Cannon-Fenske capillaries of type 100 (as well as a reference solvent) at 30°C. The set-up was a Schott Geräte CT1250 water bath plus thermocontroller including a CK100 water cooler. The electronic measurements were done with a Schott Geräte AVS350. The IV was calculated by using the Billmeyer relationship.<sup>31</sup> Two bottles per type were analysed; the results are presented as the average and the standard deviations of the two bottles.

### 2.3.3 | Molecular weight distribution

Gel permeation chromatography (GPC) measurements were performed using a Viscotek VE 2001 GPC max provided with a TDA305 Triple Detector Array. Columns used were a PSS PFG analytical linear M. PolyCal PMMA was used as calibration standard. Injection volume was 100 µL, and the oven temperature was 30°C. Samples were dissolved overnight in 3-mL HFIP with 0.02M potassium trifluoroacetate (KTFA). The resulting concentration was about 5 mg/mL. For pellet, one granule per measurement was used. For bottles, a small piece of the wall or the bottom was used. Measurements were performed in twofold.

### 2.3.4 | Differential scanning calorimetry

The rPET pellets were analysed with a Perkin Elmer DSC-8000 in twofold. Ten to 15 mg of grinded pellets were weighted into aluminium cups. The closed cups were subjected to two different temperature programmes with differ in the cooling rate either 10°C or 100°C per minute: conditioning for 5 minutes at 0°C, heating at 10°C per minute to 280°C, cooling to 0°C, conditioning for 5 minutes at 0°C, heating at 10°C per minute to 280°C. The degree of crystallisation, the melting point, and the onset of crystallisation were derived from the second heating curve after cooling at 10°C per minute, assuming a theoretical melt enthalpy for a PET crystal of 140 J/g. The glass transition temperatures were derived from the second heating run from the quickly cooled samples.

DSC measurements were also performed on fragments of the bottom parts of the PET bottles to measure the degree of crystallinity and to potentially find correlations with the ESC data. The same equipment was used. The temperature profile was stabilised at 0°C for 3 minutes and heating the material to 300°C at 10°C per minute. The degree of crystallinity was determined from the melting peak and the cold crystallisation peak according to the method of Negoro et al.<sup>32</sup>

### 2.3.5 | IR spectrophotometry

Fourier transform infrared (FT-IR) spectra of the PET bottle wall samples were obtained on a Varian Scimitar 1000 FT-IR spectrometer equipped with a DTSG detector. The measurement resolution was set at  $4\text{ cm}^{-1}$ , and the spectra were collected in the range  $4000$  to  $650\text{ cm}^{-1}$  for attenuated total reflectance (ATR) with 50 coadded scans. The sample chamber was purged by  $\text{N}_2$  gas for 10 minutes before scanning was started. ATR was performed on a PIKE MIRacle ATR equipped with a diamond w/ZnSe lens single reflection plate. The FTIR-ATR measurements were performed with three different pieces of PET bottle wall from different bottles with the same composition and averaged.

### 2.3.6 | Optical properties

#### Colour

Colour of the bottles was measured using Konica Minolta Chroma meter CR-400 according to the CIELAB method. Five bottles per type were measured. The flat surface of the lower part of the bottle was cut in pieces of  $7 \times 20\text{ cm}$ . For each bottle-wall fragment five measurements were performed. As background reference a white tile was used. This yielded three colour parameters:  $L^*$  (100 = white; 0 = black),  $a^*$  (positive = red; negative = green; 0 = grey), and  $b^*$  (positive = yellow; negative = blue; 0 = grey). These colour parameters can be combined in one number:  $\Delta E$ . It gives the total colour difference between the bottles with recycled content and the bottles with virgin PET:  $\Delta E = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$ . Since this is a calculated value, it was not considered in the correlation analysis.

#### Haze

Haze measurements were performed on the same bottle wall pieces as used for the colour measurement. Measurements were done with a BYK haze-gard plus according to norm ASTM D1003. Five bottles per type were evaluated. For each bottle wall five measurements were recorded.

#### UV-visual spectrophotometry

The UV-visual absorption spectra were measured by using a Shimadzu UV-1650PC UV-visible spectrophotometer with a Shimadzu CPS controller (six-fold cuvette holder). An 8-mm wide strip from three of the bottles' pieces used for colour and haze was folded double in the cuvette. The spectra were measured in the wavelength range from 320 to 600 nm. The thickness was measured with a Mitutoyo micrometer. The thickness normalised absorption values at 350 nm are reported.

### 2.3.7 | Mechanical properties

#### Environmental stress cracking

ESC was studied in a dedicated set-up where five bottles could be tested simultaneously under controlled pressure conditions. The set-up consists of five metal bottle holders with an inverse screw to connect the bottles. Each holder is provided with a barometer and a valve

that can be closed when the pressure in the bottles reaches the pre-set value of  $5.4 \pm 0.2\text{ bar}$ . The protocol from the International Society of beverage technologists (ISBT) was slightly adjusted (20 instead of 30 bottles were tested per type, and five bottles were tested simultaneously).<sup>33</sup> In total, 20 bottles per type were prepared for testing. Before testing, bottles were aged for 24 hours in an oven at  $50^\circ\text{C}$  and at room humidity conditions (around 50% RH). Ageing occurred between 9 and 15 days after production for bottles made of materials A, B, and 100% virgin PET. The bottles made with material C were aged 30 days after production. After ageing, bottles were conditioned at  $23^\circ\text{C}$ , 50% RH for at least 16 hours. A solution of 0.20% sodium hydroxide (NaOH) was prepared to conduct all tests. Bottles were filled with room temperature tap water till around 2 cm under the neck. Each full bottle was carefully screwed to the set-up. After all five bottles were connected, pressurised air was applied until the pressure reached  $5.4 \pm 0.2\text{ bar}$ . The valve used to pressurise the air was closed, and the barometer was checked during 5 minutes to ensure that the pre-set pressure level was maintained. After 5 minutes of holding the pressure, the water level was marked on the bottle wall. The bottle bottoms were carefully immersed in cylindrical dishes containing the NaOH solution. As the last bottle was introduced in the solution (delay around 30 seconds), the stopwatch started. The bottles, pressure, and water levels were monitored in time. In case a bottle bursts or leaks, the time to defect was noted. In case of a bottle burst, a clear explosion is heard, and an immediate drop in pressure was noted; the bottle was also immediately empty. In case of a bottle leakage, the time when the pressure starts to drop (usually happened only gradually) or the water level changes is noted. The amount of bottles bursting or leaking in the first 5 minutes, between 5 to 10 minutes and after 10 minutes, are reported.

### 2.4 | Statistical analysis

All data collected for the 13 types of bottles were studied by means of bivariate scatter plots and correlation. This statistical analysis of the data was done with the software R<sup>34</sup> with a dedicated script. Special attention was paid to the cases where a correlation was found greater than 0.8. For those pairs a linear regression analysis was performed. Some of the found correlation coefficients are fairly high (very close to 1.00), due to a small data set of 13 points.

## 3 | RESULTS

### 3.1 | Bottle properties in relation to recycled content and rPET type

#### 3.1.1 | Particle contamination

The total amount of particles in dissolved PET bottle fragments for the 13 types of PET bottles are listed in Table 1. The amount, type, and size distribution of these particles are listed in Tables SB2 and SB4. Here, only the total amount of particles is considered.

**TABLE 1** Properties dependent on recycled content: Total particle count in PPTI (single measurement), haze values, thickness corrected absorption values at 350 nm and colour parameters L\*, a\*, b\*, and  $\Delta E$  of the produced bottles, as measured/calculated for bottles with different recycled content and rPET types A, B, and C

Sample	All Particles [PPTI]	Haze [%]	$A_{350nm}/x$ [A/mm]	L*	a*	b*	$\Delta E$
Virgin PET	1525	0.7 ± 0.1	0.72 ± 0.07	95.5 ± 0.2	0.04 ± 0.04	2.4 ± 0.1	
25% A	5932	1.1 ± 0.2	0.97 ± 0.07	94.3 ± 0.3	-0.13 ± 0.05	2.6 ± 0.1	1.2
50% A	10 557	1.7 ± 0.3	1.24 ± 0.08	93.5 ± 0.4	-0.24 ± 0.04	2.6 ± 0.1	2.1
75% A	18 261	2.8 ± 0.4	1.36 ± 0.07	91.9 ± 0.6	-0.39 ± 0.06	2.4 ± 0.1	3.6
100% A	22 908	3.6 ± 0.5	1.68 ± 0.14	90.7 ± 0.9	-0.54 ± 0.08	2.7 ± 0.2	4.9
25% B	16 715	2.0 ± 0.3	0.95 ± 0.03	94.6 ± 0.3	0.04 ± 0.07	3.3 ± 0.2	1.5
50% B	29 543	3.7 ± 0.6	1.29 ± 0.08	93.6 ± 0.4	-0.22 ± 0.06	4.1 ± 0.3	2.5
75% B	43 779	5.0 ± 0.6	1.42 ± 0.10	92.8 ± 0.6	-0.45 ± 0.09	4.9 ± 0.4	3.7
100% B	62 572	6.6 ± 1.0	1.75 ± 0.04	91.7 ± 0.7	-0.72 ± 0.11	5.8 ± 0.5	5.2
25% C	13 387	1.8 ± 0.2	1.02 ± 0.03	94.3 ± 0.4	-0.15 ± 0.08	3.2 ± 0.2	1.5
50% C	24 877	3.3 ± 0.4	1.30 ± 0.09	92.9 ± 0.5	-0.30 ± 0.11	3.9 ± 0.3	3.0
75% C	35 452	4.7 ± 0.8	1.41 ± 0.09	92.0 ± 0.8	-0.38 ± 0.09	4.2 ± 0.4	4.0
100% C	51 303	6.2 ± 0.9	1.68 ± 0.06	91.1 ± 0.9	-0.48 ± 0.07	4.5 ± 0.4	4.9

Abbreviations: PET, poly (ethylene terephthalate); PPTI, particles per 10 000 images.

The lowest amount of particles is found in solutions made of virgin PET bottles, around 1500 PPTI. This figure is much lower than the results obtained on bottles made of 100% A, B, and C rPET types. These are, namely, 14, 40, or 30 times higher than the amount obtained for bottles made of virgin PET. The amount of particles in dissolved bottles is in most cases higher than in the dissolved pellets where the bottles were made from, see Table SB2 (Supporting Information).

For bottles with recycled content, more than 70% of the counted particles belong to the smallest size category of 0 to 3  $\mu\text{m}$ , irrespective of rPET type and recycled content. Furthermore, more than 90% of all detected particles are smaller than 5  $\mu\text{m}$ . This was in contrast with the particles found in bottles made of virgin materials. Here, around 50%

are in the category 0 to 3  $\mu\text{m}$ , and in total, around 75% are smaller than 5  $\mu\text{m}$ .

The total PPTI found in solutions of PET bottles increased significantly with the recycled content for all rPET types. Statistical analysis showed that the total amount of particles increased linearly with the recycled content, and the slopes are different for all three types of rPET. Linear fit results are shown in Table 2. The intercept is zero due to the relative large error in the initial value. The data were fitted in three groups, excluding the reference value for virgin PET, in three curves with a coefficient of determination of 0.9986. Type A rPET showed the smallest amount of PPTI and the least steep slope whilst the steepest slope was found for rPET B. The amount of particles in the rPET bottles was found to depend both on the recycled content and

**TABLE 2** Results of the linear regression analysis done for some of the studied properties vs rPET content, slope, and intercept per rPET type with its correspondent error are reported

Studied Property		rPET Type A	rPET Type B	rPET Type C	Multiple R <sup>2</sup>
All particles	Slope	23 000 ± 3100	61 000 ± 3100	50 000 ± 3100	.99
	Intercept	0	0	0	
Haze	Slope	3.5 ± 0.2	6.0 ± 0.2	5.7 ± 0.2	.99
	Intercept	0.2 ± 0.1	0.6 ± 0.1	0.4 ± 0.1	
UV A350	One slope	0.92 ± 0.06			.99
	One intercept	0.77 ± 0.04			
Colour L*	One slope	-4.3 ± 0.2			1.00
	Intercept	95.3 ± 0.2	95.9 ± 0.2	95.3 ± 0.2	
Colour b*	Slope	0	3.4 ± 0.2	1.7 ± 0.2	.99
	Intercept	2.6 ± 0.2	2.4 ± 0.2	2.9 ± 0.2	
$\Delta E$	One slope		4.9 ± 0.2		.98
	One intercept		0		

Abbreviation: rPET, recycled poly (ethylene terephthalate).



the type of rPET used for bottle production. When more particles are present in the pellet before bottle production, then the slope is steeper.

### 3.1.2 | Optical properties

Four out of the five studied optical properties were found to clearly depend on the recycled content and the type of rPET. These are haze, colour  $L^*$  (lightness), and colour  $b^*$  (yellowness) according to the CIELAB method<sup>7</sup> and the thickness corrected absorption in the UV-visual spectrum at the wavelength of 350 nm. This was less obvious for the colour parameter  $a^*$  (green-red scale). The measured data are shown in Table 1.

#### Haze

The haziness of PET bottles increases linearly with the recycled content. The type of rPET used has a large influence on the measured haze of the bottles. A very good correlation was found between the haze of the bottles and the recycled content of the bottles, the fit parameters for the linear regression are given in Table 2. Type A rPET showed less increase in haze as given by the less steep slope when compared with B and C rPET types, these show comparable slopes. The measured haze values varied between 0.7% for virgin PET bottles and 6.6% for bottles made with 100% B rPET. The association of plastic recyclers (APR) indicates that a transmission haze value of 9.5% should not be a problem for 3-mm thick plate samples made of recycled or virgin pellets.<sup>35</sup> The bottles studied here are also measured in transmission haze but have wall thickness of around 200  $\mu\text{m}$ . These results cannot be compared directly since there is no known relationship between haze and thickness. Haze is a bulk property and therefore thickness play an important role in the measured value. To our knowledge, there is no haze specification for bottle walls.

#### Colour

Similar as with the haze, the colour of the bottles also varies with the recycled content and is dependent on the type of rPET used.  $L^*$  (lightness) of the bottles decreases with the recycled content. It does not seem to depend on the type of rPET, meaning that bottles will always become darker when rPET is added, independently of the quality of the added rPET. The minimum value obtained for  $L^*$  in this study is 91 for bottles containing 100% recycled content. This figure is far above the 82 value given by the APR for 3-mm thick plaques of clear PET.<sup>35</sup> As in the case of haze, introducing recycled content decreases the lightness of the bottle walls. However, for  $L^*$  values, the decrease depends only on the recycled content added and not on the type of rPET (Table 2).

PET bottles with recycled content have higher positive  $b^*$  values than virgin PET bottles (see Table 1), implying that they are more yellow. In general, the amount of yellowing increases with the recycled content. But the yellowing depends also on the type of rPET used. Regression analysis shows the slope for each type of rPET is completely different (see Table 2). For rPET type A, the  $b^*$  parameter does not change with rPET content (slope is 0), whereas for the rPET types B and C, the  $b^*$  values increase strongly to values as high as 6.

These different slopes between type A and B, C relate to the use of a blue colorant by the producer of type A, which masks the yellowing. The  $a^*$  values of the PET bottles vary in values close to zero and are not further analysed. The total colour difference  $\Delta E$  increases linearly with the recycled content, independent of the type of rPET added.

#### Ultraviolet-visual spectrophotometry

The thickness-corrected absorption value at 350 nm of the bottle wall fragments correlates very good with the percentage of added rPET, for all three types of rPET. This relation is independent of the rPET type used. Other wavelengths close to 350 nm show the same behaviour, namely, 400 and 338 nm. Thickness-corrected absorption data for 350 nm is shown in Table 1. These results on PET bottle walls are in agreement with previous reported data on PET sheets.<sup>8,18</sup> This fundamental relationship applies to clean polymeric materials. In practice, PET and rPET suppliers use additives to influence the colour of the pellets, and this changes the final colour of the bottle walls as well. This will hamper the practical use of this correlation to predict the recycled content of PET bottles in the market.

### 3.1.3 | IV and molecular weight

IV is a prime market requirement for bottle grade rPET. According to literature, the IV of the pellet has to be between 0.73 and 0.8 dL/g for carbonated drink bottles applications.<sup>36</sup> However, in the last years, requirements have shifted towards higher values. Bottle producers require higher IV values for carbonated drinks or beer in Europe, above 0.80 dL/g and even higher in tropical areas. In Table 3, the IV results for pellets and bottles made of 100% rPET and virgin pellets and bottles with 25% rPET content are shown. The IV of pellets is close to the literature specifications ( $IV > 0.73$  dL/g).<sup>36</sup> The virgin pellet used in this study had an IV value below 0.8 dL/g. The highest IV values were found for rPET A, and the lowest values were found for rPET B.

During the processing of the pellets, the materials were exposed to 290°C for 33 seconds during injection moulding of the preforms, and afterwards, these preforms were reheated to temperatures around 100°C to 115°C for subsequent bottle blowing. Exposure of PET to high temperatures results in a reduction in molecular weight by thermomechanical degradation, resulting in a drop in IV. A reduction in IV of around 9% to 14% is observed for bottles containing 100%

**TABLE 3** Intrinsic viscosity in dL/g as measured according to ASTM D4603-03 for pellets, bottles containing 100% of the pellet and bottles containing 25% of the corresponding rPET pellets

Material	Pellets	Bottles Containing 100% Pellets	Bottles Containing 25% rPET
Virgin PET	0.78 ± 0.01	0.71 ± 0.06	NA
rPET A	0.81 ± 0.02	0.73 ± 0.01	0.72 ± 0.01
rPET B	0.73 ± 0.01	0.63 ± 0.01	0.66 ± 0.04
rPET C	0.77 ± 0.02	0.66 ± 0.02	0.76 ± 0.01

Abbreviations: NA, not applicable; rPET, recycled poly (ethylene terephthalate).

of the pellet materials. Similar values (drop in viscosity of around 14%) have been found when reprocessing dried PET materials at 270°C.<sup>22</sup> This drop in viscosity is extremely high when compared with commercial conditions and most likely related to the long exposure times to elevated temperatures in the small scale production facility.

Bottles containing 25% rPET were also analysed; they were chosen because of their differences in reheating temperatures: Bottles containing 25% rPET type C were reheated at a lower temperature (around 100°C) than the rest, and the effect of this reheating step can be clearly seen in the high values of the IV comparable with the original pellet. This confirms the large influence of the processing conditions on the IV drop. Preform reheating temperatures are shown in Table 4.

Several attempts were made to study molecular weights of the bottles by GPC techniques. However, the deviation found in repeated measurements for the molecular weights (Mw, Mn, and Mz) and the fractions of molecules smaller than 10 000 or 20 000 g/mol was larger than the difference between the values for different samples. For all bottles, average molecular weights (Mw) were 52 000 ± 5000 g/mol, Mn values were 26 000 ± 3000 g/mol, Mz values were 87 000 ± 12 000 g/mol, and the percentage fraction below 20 000 was 21 ± 2% and below 10 000 was 6 ± 1% independent of the recycled content or the type of material used to produce the bottles, so also for virgin materials. The large standard deviation found in the measurements probably relates to the small sample size used in GPC (around 15 mg), which could result in a large error when comparing similar samples.

### 3.1.4 | Differential scanning calorimetry

The thermal behaviours of the three types of rPET pellets are all very similar, see Table SD1 (Supporting Information). The thermal behaviour of the bottom parts of the PET bottles does reveal a few differences, see Table SD2 (Supporting Information). The degrees of crystallisation

are listed in Table 4, and it varies between 23% and 46%. For type C rPET, the degree of crystallisation of the bottle bottom does not vary with the recycled content. However, for type A at 50% recycled content, for type B at 50%, and 75% recycled content, the degree of crystallisation is clearly reduced, and cold crystallisation is observed.

### 3.1.5 | IR spectrophotometry

Subtracted IR spectra between 750 and 700 cm<sup>-1</sup> of the PET bottle walls are shown in Figure SE1 (Supporting Information). This region shows the rocking vibration of the methylene moieties in the polymer backbone. A larger absorption is indicative for a more crystalline structure.<sup>18</sup> All three types of rPET have different influences on this IR absorption peak. For type A rPET, this absorption is reduced only for 50% recycled content. For type B rPET, this absorption is slightly reduced for 50% recycled content and increased for 25% and 100% recycled content. For type C rPET, the absorbance is reduced for 25%, 50%, and 75% recycled content and then raised for 100% recycled content. These results suggest that the type of rPET and the recycled content have a subtle, inconsistent influence on the degree of crystallinity of PET bottle walls.

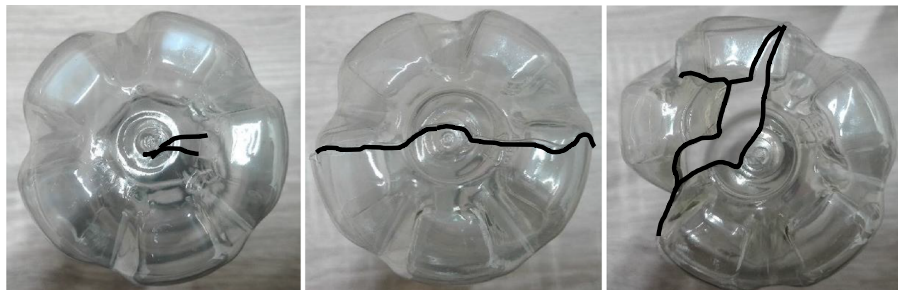
### 3.1.6 | Environmental stress cracking

ESC was evaluated for 20 bottles of each material type. Two different types of failures were discerned. A leak is a very small hole close to the injection point, almost not perceptible to the bare eye. A burst resulted in either tearing the bottle in half or bursting it completely open. The types of damage observed are shown in Figure 1. In Table 4, the amount of bottles, which burst within the first 5 minutes of the test, the amount of bottles that burst or leak between 5 and 10 minutes, and the amount of bottles, which leak after 10 minutes, are

**TABLE 4** Amount of bottles successfully tested in ESC tests, expressed as the amount of bottles burst in the first 5 min, between 5 and 10 min, and after 10 min, complemented with the temperature of the preform and the degree of crystallinity of the bottle bottom according to DSC

Sample	Total Bottles Burst in First 5 min	Total Bottles Burst/Leak Between 5 and 10 min	Total Bottles Burst/Leak After 10 min	Total Bottles not Tested	Temperature Preform, °C	Degree of Crystallinity of the Bottle Bottom, %
Virgin PET	16	0	1	3	112	38
25% A	15	0	3	2	109	41
50% A	6	2	9	3	110	29
75% A	3	5	12	0	110	42
100% A	5	5	9	1	109	39
25% B	18	0	1	1	113	46
50% B	13	3	1	3	116	31
75% B	19	1	0	0	114	23
100% B	17	2	0	1	106	40
25% C	10	2	8	0	100	38
50% C	19	1	0	0	115	40
75% C	17	1	2	0	111	38
100% C	16	2	2	0	110	41

Abbreviations: ESC, environmental stress cracking; PET, poly (ethylene terephthalate).



**FIGURE 1** Identified types of damage after the environmental stress cracking test. From left to right: leak (puncture or a small rupture), burst when the bottom is split in half and a complete burst. Black lines were drawn to accentuate the defects

reported. Some of the bottles (a maximum of three for some types) leaked already when tightening them to the experimental set-up or during the pressure stabilisation time; therefore, the amount of bottles that could not be tested is also reported. Within the first 5 minutes, no leaks were registered, all bottles that fail are burst. In contrast, after 10 minutes, most bottles begin slowly to leak, but bursts were also observed. Detailed results of the corresponding times to failure are given Table SC1 (Supporting Information).

The vast majority of bottles (16 of the tested 17) made from virgin PET (reference material) burst within the first 5 minutes, and only one bottle resisted more than 10 minutes. For all bottles containing rPET, more than 70% of the tested bottles burst in the first 5 minutes with exception of four bottle types: bottles containing 50%, 75%, and 100% of rPET type A, and the bottles containing 25% rPET type C. For these four bottle types, not only the amount of bottles burst in the first 5 minutes is low but also the amount of bottles leaking after 10 minutes is much higher than for all the other bottle types (eight to 12 bottles in comparison with one to three for the other bottle types). These results do not depend on the recycled content.

The results for bottles containing more than 50% rPET type A are surprising, less than 40% of the bottles burst in the first minutes, and more than 40% did not leak after 10 minutes of testing, indicating a good ESC resistance. Results on 100% rPET type A bottles compared with virgin materials indicate that rPET type A is inherently better resistant against ESC than the virgin material used as reference in this study. Additionally, the results suggest that at least 50% of this high quality rPET material is needed to get improved stress cracking resistance since bottles containing 25% still show lower performance.

Process conditions during the bottle production, namely, preform reheating temperatures can explain the difference encountered for bottles containing 25% rPET type C. These bottles were produced at lower temperatures than the rest of the bottles (100°C instead of 110°C as shown in the last column of Table 4).

### 3.2 | 3.2 Correlations between bottle properties and material parameters

The statistical analysis of the generated data for the various PET bottles gives three important correlations between the optical properties

haze, colours  $L^*$  and  $b^*$ , and particle contamination of PET bottles. These are presented and analysed in detail in this section. Further, a first indication of a possible correlation between ESC and IV was found. No other correlations were found with for instance the degree of crystallinity from DSC or the absorption area from IR and the occurrence of ESC.

#### 3.2.1 | Haze and particle contamination

A single line correlation was found between the particle contamination in PET bottles and the haze of PET bottle walls. This indicates that the haziness of PET bottles depends on the amount of particles present in the bottle wall. Although the amount of particles is different for each rPET type, their effect on haze is apparently the same. Linear regression parameters are shown in Figure 2.

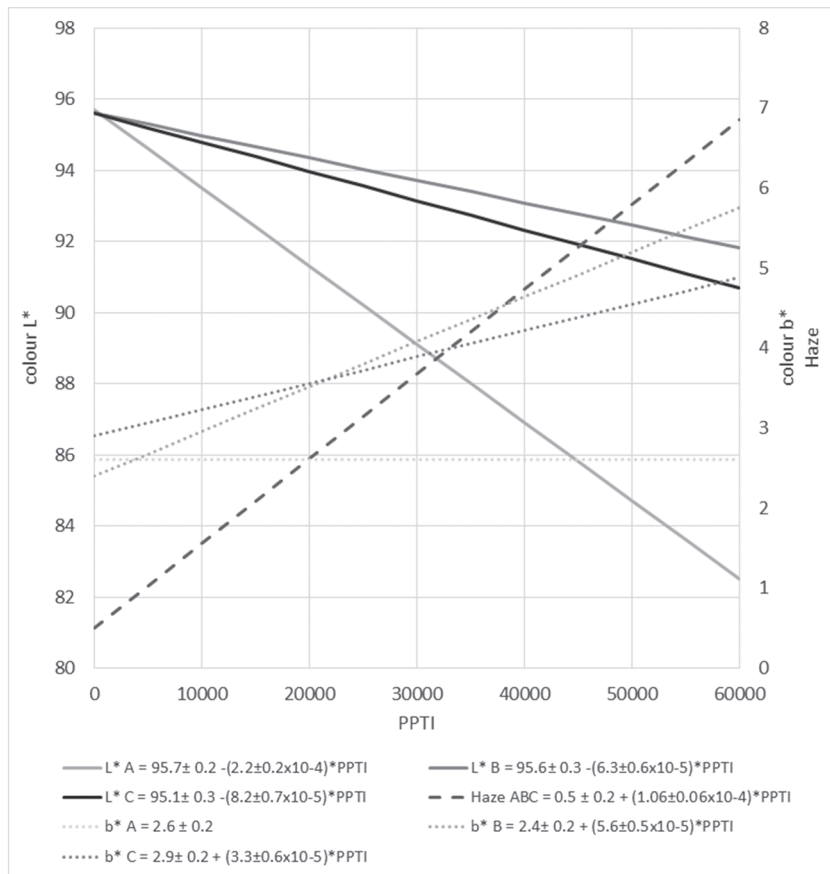
#### 3.2.2 | Discolouration ( $L^*$ and $b^*$ ) and particle contamination

A good correlation was also found between lightness ( $L^*$ ) and yellowing ( $b^*$ ) of the PET bottles with the amount of particles present in the walls of PET bottles. Here, no single line could be used to fit all three types of rPET, meaning that the change in colour of the bottles is dependent not only on the amount of particles present but also on the type of rPET used.

Colour parameter  $L^*$  was found to decrease with increasing amounts of particles. For type A rPET, the  $L^*$  parameter decreased the most with PPTI, whereas this decrease was substantially less for the other types of rPET B and C. This most probably relates to the presence of additives to mask the yellowing of rPET. The fact that both rPET types B and C have slightly different slopes could indicate that there is a difference in the type of particles present in both types of rPET and that, apparently, the particles present in type B decrease the  $L^*$  parameter to a lower extent. The lightness ( $L^*$ ) decreases for all rPET types with increasing levels of particles in the PET bottles, but other factors such as additives and type of present particles also have a distinctive influence.

Colour parameter  $b^*$  also showed a different trend for type A than for types B and C. For type A, the  $b^*$  parameter did not change with





**FIGURE 2** Trends for the optical properties vs the total amount of particles (PPTI) from the linear regression analysis. Haze is reported as one fit for all data,  $R^2 = .96$ . Colour  $L^*$  ( $R^2 = 1.00$ ) and colour  $b^*$  ( $R^2 = .99$ ) as three different curves. Equations per property are shown in the legend of the graph

amount of particles, slope was zero. For types B and C, the  $b^*$  parameter clearly increased with different slopes with amount of particles. This can be explained by the use of “blue agents” in type A and not in types B and C.  $b^*$  values of pellets were different when received, they are reported in Table SB3 (Supporting Information). Material type B was already yellow before processing in bottles; the process only enhances this yellow colour and results in the most yellow coloured bottles. In contrast, material A had similar  $b^*$  values as the reference material. After mixing these two materials,  $b^*$  parameter does not change.

### 3.2.3 | ESC vs IV

Process conditions are critical for properties such as IV and ESC as shown in Tables 3 and 4. The drop in IV for bottles containing 25% rPET type C is lower than for all the other bottles containing the same rPET type. Furthermore, bottles containing 100% rPET type A show higher IV when compared with 100% rPET bottles with rPET types B and C. Both observations indicate that ESC could be related to the higher IV after processing.

A general trend could be found between the IV of the PET bottles and the share of bottles that leaked after 10 minutes. The higher the IV of the PET bottle, the larger the amount of bottles that will resist more than 10 minutes in the ESC test. The data are insufficient to determine a proper correlation, but the trend is clear.

## 4 | DISCUSSION

### 4.1 | Impact of the bottle production method on the bottle properties

The PET bottles for this study were produced in a small scale production facility, which resulted in longer exposure times of the preforms to elevated temperatures than what is common in industrial large scale production facilities. Processing will impact the properties of PET bottles related to thermally induced degradation reactions, which become apparent as yellowing and lower IV's. Therefore, the values found for the colour parameter  $b^*$  in relation to recycled content are likely to be an overestimation as compared with the industrial reality. Also, the reduction in IV from pellets to bottles measured in this study is likely to be an overestimation and a worst case scenario. This will probably influence the measured mechanical properties as shown. Nevertheless, optical properties (haze and  $L^*$ ) and measured particle contamination are not sensitive to the temperature and hence the relationships found will not be influenced by the production method.

### 4.2 | The relevance of particle contamination for PET bottles with recycled content

The particles in solution measurements proved that rPET contains substantially more particles than virgin PET. The data reveal that

higher levels of particle contamination can be found in rPET originating from cocollection systems (types B and C) compared with monocollection systems for bottles (type A). The origin of the contaminants is further discussed in a separate paper.<sup>37</sup>

Haze correlates linearly with the particle contamination, more particles imply more haze. For the colour parameters  $L^*$  and  $b^*$ , the relationships are more complex; here, not only particle contamination but also other factors like additives and, namely, colour masking agents influence the relationship. If blue colouring is added to type A rPET, then this will explain not only the lack of change of  $b^*$  with rPET content and particle content but also the strong decrease of  $L^*$  when increasing amount of particles.

For rPET types B and C, increase in  $b^*$  with particle content might be explained by two mechanisms. Either the particles themselves give colour to the rPET (and therefore  $b^*$  increases with increasing particle content) or the particles might catalyse chemical reactions, which would enhance the formation of conjugated aromatic systems with a yellow colour, also resulting in increasing  $b^*$ .

The particle contamination might even have an indirect impact on the molecular weight of the rPET, since various polymeric contaminants (PS, PP, EVOH, and PVC) have been found to retard the restoration of chain lengths in the SSP process<sup>13</sup> and hence contribute to lower IV's of the rPET.

Particles in solution measurements are a valuable tool to screen particle contamination in rPET. However, the origin of the particles is not always clear. Because of the direct linear relationship with haze, a haze measurement would even suffice as quality screening method to determine the amount of particles present. The acceptance of a hazy bottle depends on the colour of the beverage and the marketing strategy of beverage company. Haziness could be more accepted by the consumers if they are aware that it indicates the use of recycled content in the bottles. Colour and haze are marketing aspects and are not always regarded negatively. Colour  $b^*$ , for example, can also be largely influenced with additives.

### 4.3 | Impact of the IV on ESC

The occurrence of ESC in PET bottles could not be correlated to the presence of rPET. Hence, the opinion of several representatives of beverage companies that using rPET increases the chance that ESC occurs could not be confirmed. However, a trend with the process conditions and IV of the PET in the bottles was found. In case the applied types of rPET have lower IV's, the chance on ESC occurring grows. IV of rPET is primarily determined by SSP processing parameters and can be "tuned" during rPET production. However, the presence of polymeric contaminants that hinder and or retard the restoration of chain lengths in the SSP process, could influence the maximum IV to achieve, and therefore increase the chance of ESC. It is certain that high IV values decrease the chance of mechanical failure in the first 5 minutes of the ESC tests of PET bottles.

## 5 | CONCLUSIONS

The influence of recycled content and type of rPET on the particle contamination, IV, optical properties, and ESC of PET bottles has been systematically studied. The particle contamination increases with the recycled content.

A direct linear relationship was found between the haziness and the particle contamination; the more contamination, the more hazy the bottle. Colour parameters  $L^*$  and  $b^*$  were found to linearly correlate with the recycled content, but the slope of the correlation was different for different types of rPET. In general, the bottles become greyer and yellower with increasing levels of recycled content. Most likely, not only the particle contamination but also other factors such as the addition of blue colorants and the type of particle contaminants also play an important role in these colour parameters. The more recycled content added, the larger the observed difference in colour with respect to virgin materials, independently of the rPET type added.

No relationship was found between the recycled content and the occurrence of ESC of PET bottles. However, a general trend was found between the chance that ESC occurs and the IV of the PET bottle, the higher the IV, the smaller the chance of ESC occurring.

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### REFERENCES

1. PETCore. 2017; <https://www.petcore-europe.org/news-events/110-pet-collection-recycling-rates-europe-significantly-2016.html>.

2. Welle F. Twenty years of PET bottle to bottle recycling—an overview. *Resour Conserv Recy*. 2011;55(11):865-875.
3. Simon B, Ben Amor M, Foldenyi R. Life cycle impact assessment of beverage packaging systems: focus on the collection of post-consumer bottles. *J Clean Prod*. 2016;112:238-248.
4. Chilton T, Burnley S, Nesaratnam S. A life cycle assessment of the closed-loop recycling and thermal recovery of post-consumer PET. *Resour Conserv Recy*. 2010;54(12):1241-1249.
5. Clark D. Processing PCR: How it's done at a leading PET bottle maker. *Plastics Technology*. 2010;56(2):18-20.
6. McKeen L. Introduction to the physical, mechanical, and thermal properties of plastics and elastomers. *The Effect of Sterilization on Plastics and Elastomers*. Third ed 2012.
7. X-rite. A Guide to Understanding Color. 2016; [https://www.xrite.com/-/media/xrite/files/whitepaper\\_pdfs/I10-001\\_a\\_guide\\_to\\_understanding\\_color\\_communication/I10-001\\_understand\\_color\\_en.pdf](https://www.xrite.com/-/media/xrite/files/whitepaper_pdfs/I10-001_a_guide_to_understanding_color_communication/I10-001_understand_color_en.pdf). Accessed 07 March 2019.
8. Curtzwiler G, Vorst K, Danes JE, Auras R, Singh J. Effect of recycled poly (ethylene terephthalate) content on properties of extruded poly (ethylene terephthalate) sheets. *Journal of Plastic Film & Sheeting*. 2011;27(1-2):65-86.
9. Misra E, Basavarajaiah S, Kumar KR, Ravi P. Effect of recycling on the properties of poly (ethylene terephthalate) films. *Polym Int*. 2000;49(12):1636-1640.
10. Thoden van Velzen EU, Brouwer MT, Stärker C, Welle F. The effect of recycled content and rPET quality on the properties of PET bottles, part II: migration. *Submitted to Packag Technol Sci*. 2019.
11. Jabarin SA. Strain-induced crystallization of poly (ethylene-terephthalate). *Polym Eng Sci*. 1992;32(18):1341-1349.
12. Dvorak R, Kosior E, Fletcher J. Improving food grade rPET quality for use in UK packaging. WRAP;2013. *Final report*.
13. Thoden van Velzen EU, Brouwer MT, Molenveld K. *Technical quality of rPET that can be obtained from Dutch PET bottles that have been collected, sorted and mechanically recycled in different manners*. Wageningen: Wageningen Food & Biobased Research; 2016 Report number 1661.
14. Snell H, Nassour A, Nelles M. Qualitative comparison of polyethylene terephthalate flakes from various collection systems in Germany. *Waste Manage Res*. 2017;35(2):163-171.
15. Alsewailam FD, Alrefaie JK. Effect of contaminants and processing regime on the mechanical properties and moldability of postconsumer polyethylene terephthalate bottles. *Waste Manage*. 2018;81:88-93.
16. Gebben B. Less fiber breaks with the micro scout. *Chemical Fibers International*. 2002;52.
17. MacDonald WA. New advances in poly (ethylene terephthalate) polymerization and degradation. *Polym Int*. 2002;51:923-930.
18. Curtzwiler GW, Williams EB, Maples AL, et al. Ultraviolet protection of recycled poly (ethylene terephthalate). *J Appl Polym Sci*. 2017;134(32):45181.
19. Lecomte HA, Liggat JJ. Degradation mechanism of diethylene glycol units in a terephthalate polymer. *Polym Deg Stab*. 2006;91:681-689.
20. Rieckmann T, Besse K, Frei F, Völker S. Quantification of colour formation in PET depending on SSP residence time, temperature, and oxygen concentration. *Macromolecular Symposia*. 2013;333(1):162-171.
21. Momany T. *The Impact of Recycled PET Content on PET Hot-Fill Containers*. Holland, USA: PTI: Plastic Technologies, Inc. (PTI); 2016.
22. Voyles J. *The Impact of Repeated PET Melt Cycles on Color*. Holland, USA: PTI: Plastic Technologies, Inc. (PTI); 2018.
23. Masmoudi F, Fenouillot F, Mehri A, Jaziri M, Ammar E. Characterization and quality assessment of recycled post-consumption poly (ethylene terephthalate) (PET). *Environ Sci Poll Res Int*. 2018;25(23):23307-23314.
24. Ciolacu CFL, Choudhury NR, Dutta NK. Colour formation in poly (ethylene terephthalate) during melt processing. *Polym Deg Stab*. 2006;91(4):875-885.
25. La Mantia FP, Vinci M. Recycling poly (ethyleneterephthalate). *Polym Deg Stab*. 1994;45:121-125.
26. Badia JD, Vilaplana F, Karlsson S, Ribes-Greus A. Thermal analysis as a quality tool for assessing the influence of thermo-mechanical degradation on recycled poly (ethylene terephthalate). *Polymer Testing*. 2009;28(2):169-175.
27. Demirel B, Daver F. The effects on the properties of PET bottles of changes to bottle-base geometry. *J Appl Polym Sci*. 2009;114(6):3811-3818.
28. Prasath RGR, Newton T, Danyluk S. Stress monitoring of PET beverage bottles by digital photoelasticity. *Manufacturing Letters 15PA*. 2018;9-13.
29. Morrison ED, Malvey MW, Johnson RD, Anacker JL, Brown KA. Effect of chemical environments on stress cracking of poly (ethylene terephthalate) beverage bottles. *Polymer Testing*. 2008;27(6):660-666.
30. Coriolani C, Licciardello F, Muratore G. Stress-cracking of pet bottle as an unpredictable factor limiting the commercial life of carbonated soft drinks. *Italian Journal of Food Science*. 2012;24(4):170-173.
31. Negoro T, Thodsaratpreeyakul W, Takada Y, Thumsorn S, Inoya H, Hamada H. Role of crystallinity on moisture absorption and mechanical performance of recycled PET compounds. *Energy Procedia*. 2016;89:323-327.
32. Billmeyer FW. Methods for estimating intrinsic viscosity. *J Polym Sci*. 1949;4(1):83-86.
33. ISBT. *Test method for evaluating the relative stress crack resistance of poly (ethylene terephthalate) carbonated soft drink bottles*. Vol PTC-Stress Crack Test Methods 031207: "International Society of beverage technologists; 2007.
34. The R Foundation. <https://www.r-project.org/>. Accessed 17-12-2018, 2018.
35. APR. PET heat history and discoloration evaluation. Vol PET-S-03: The Association of Plastic Recyclers; 2018.
36. Awaja F, Pavel D. Recycling of PET. *Eur Polym J*. 2005;41(7):1453-1477.
37. Brouwer MT, Alvarado Chacon F, Thoden van Velzen EU. Effect of recycled content and rPET quality on the properties of PET bottles, part III: modelling of repetitive recycling. *Accepted in Packag Technol Sci*. 2019.

## SUPPORTING INFORMATION

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

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