Received: 5 November 2021 | Revised: 20 December 2021 | Accepted: 21 December 2021
DOI: 10.1002/pts.2633

RESEARCH ARTICLE

Packaging Technology and Science
An International Journal
WILEY

Effect of poly lactic acid trays on the optical and thermal properties of recycled poly (ethylene terephthalate)

Eggo Ulphard Thoden van Velzen[1] | **Sharon Chu**[1] | **Karin Molenveld**[1] | **Vladislav Jašo**[2]

[1]Wageningen Food & Biobased Research, Wageningen University & Research, Wageningen, The Netherlands

[2]Total Corbion PLA BV, Gorinchem, The Netherlands

Correspondence
Eggo Ulphard Thoden van Velzen, Wageningen Food & Biobased Research, Wageningen University & Research, Bornse Weilanden 9, 6709 WG Wageningen, The Netherlands.
Email: ulphard.thodenvanvelzen@wur.nl

Funding information
Total Corbion PLA

[Correction added on 02 February 2022, after first online publication: The author's family name, Thoden van Velzen, has been corrected on the running head and citation.]

Abstract

To progress towards a more sustainable plastic system, multiple interventions are required, including the decoupling from fossil feedstock. Biobased plastics therefore have to be integrated in plastic waste management systems. This should, however, not hamper the performance of current recycling systems. Several studies have previously suggested that the uptake of poly lactic acid (PLA) in this system would endanger poly (ethylene terephthalate) (PET) recycling. This study reports the estimated concentration of PLA in recycled PET and the effect of the presence of this impurity on the optical and thermal properties of recycled PET. The current concentrations of PLA in recycled PET in the Netherlands were modelled to vary between 0% and 0.019%. When the PLA consumption rises, the concentration in recycled PET can be kept below 1% with NIR technologies. The impact of 0.1 to 1% PLA in recycled PET on the optical and thermal properties was negligible. Conversely, the negative impact of 0.1% PVC was substantial. Also the impact of 0.1% EVOH on recycled PET was studied and like PLA found to be limited. This study therefore contravenes previous studies on the impact of PLA on the quality of recycled PET. The difference between this study and the previous studies is that within this study, recycled PET has been processed in agreement with industrial methods. Therefore, in case the sorting and recycling facilities maintain their current careful operation, no negative impact of PLA on PET recycling can be foreseen, and further integration of biobased plastics in the plastic waste management system can be pursued.

KEYWORDS
impurities, mechanical recycling, optical properties, poly (ethylene terephthalate), poly lactic acid

1 | INTRODUCTION

The European Union strives towards a circular economy to reduce the use of resources and to minimise the generation of wastes.[1] Plastics are in the focus point of attention, since they are currently least circular; hence, using large amounts of crude oil, and after use they are

---

Abbreviations: DKR, *Deutsche Kreislauf und Recycling Gesellschaft*; DRS, deposit refund system; DSC, differential scanning calorimetry; EVOH, poly ethylene-co-vinyl alcohol; IV, intrinsic viscometry; LWP, lightweight packaging waste; PET, poly (ethylene terephthalate); PLA, poly lactic acid; PVC, poly vinyl chloride; rPET, recycled poly (ethylene terephthalate); SSP, solid-state processing.

either incinerated, landfilled, recycled or leaked into nature.[2] Multiple interventions are required to progress towards a more circular economy for plastics. We need to redesign plastic articles to make them recyclable and to reduce the environmental impact of their production, to collect more plastic waste, to improve sorting technologies, to improve on and develop new recycling technologies and to divert from fossil feedstock. These interventions have to be aligned to achieve synergy.[3] Unfortunately some of the proposed interventions can potentially also counteract each other, creating dilemmas.[4] One of these potentially counteractive interventions is the inclusion of biobased plastics within the recycling system for plastics. Chemically identical biobased polymers (named drop ins) are accepted, but new polymers are often considered to jeopardise the current recycling system. Newly introduced polymers are often not present in sufficient volumes to make it financially attractive for sorting companies to create a separate sorted product for them; hence, these are added to the sorting residues and will also be present in other sorted products as sorting mistakes. These faulty sorted plastic objects made from these new bio-polymers can end up in the recycled plastics and potentially negatively affect the quality of recycled plastics. This is a well-known lock-in of the recycling system. This lock-in is currently used as argument against the introduction of PLA-based packages on the market.[5] This article studies the validity of that argument.

This research question is important, since the diversion from fossil oil feedstock is necessary to lower the environmental impacts (and especially the greenhouse gas emissions) from the plastic value chain.[2,6] In principle, two types of alternative feedstocks are foreseen: recycled plastic and renewable feedstock. The recycling system for plastics is currently still immature. Even in Europe, only 35% of the plastic packages are separately collected and only 5% is used to make new packages.[7] Since recycling systems will always have losses, both in respect to the quantity of material that is recycled[8] and in respect to the quality of material,[9] new feedstock is always required to replenish the losses in case we want to maintain our level of consumption. This new feedstock would ideally be renewable materials with lower environmental impacts. PLA is a renewable plastic that is currently available on a large scale and its application for multiple packaging applications could in principle result in lower environmental impacts such as greenhouse gas emissions in comparison to fossil-based packages.[10–13] These lower environmental impacts are attained in case the PLA packages have the same weight as the fossil based packages, do maintain the same level of product losses as the fossil based packages and the PLA packages would be mechanically recycled.[10–13] To enable the recycling of PLA packages, the collection and recycling system for lightweight packaging waste (LWP) would need to be altered, and PLA packages could be sorted out as a separate plastic waste fraction[14,15] and recycled separately.[16–18] Its application should, however, not hamper the recycling of other packages.

Multiple articles have stated that the inclusion of PLA packages in the LWP recycling system will imperil the recycling of PET bottles.[16,19,20] The reasoning is that the expected concentration of PLA in the PET bottle feedstock is approaching the threshold values that have been reported to elicit a negative effect on the quality of the recycled PET.[19] The expected concentration of PLA in PET bottle feedstocks is strongly dependent on the consumption levels, the collection and the sorting methods and hence can vary strongly between regions.

Multiple threshold values have been reported at which the presence of PLA in recycled PET would affect the quality of recycled PET in a negative manner, yielding varying limits that range from 0.1% to 5%. Alaerts et al. have presented an overview of these studies.[19] The four previous empirical studies used different materials, concentration ranges, processing methods, analysis methods and drew different conclusions. In 2012, La Mantia et al. dried PET and PLA bottle scraps, mixed them with a Brabender kneader and compression moulded samples out of it.[21] They reported that the non-isothermal flow of rPET is already negatively influenced by 0.1% PLA.[21] In 2016, McLaughlin and Ghita mixed virgin PET and virgin PLA in an injection moulding machine and reported that the mechanical properties were negatively influenced by 2% PLA.[22] In 2020, Gere and Czigany studied the compatibilisation of virgin PET and PLA in concentrations of 15% and higher, by drying, twin screw extrusion, drying and injection moulding.[23] Hardly surprising, they found negative effects of the presence of PLA on the mechanical properties at these high concentrations. In 2021, Aldas et al. compounded mixtures of virgin PET and PLA in a twin-screw extruder and injection moulded samples from it.[24] They revealed that PET and PLA are partially miscible at PLA concentrations below 10%. Additionally, they did notice a reduction in the Young's modulus, a limited reduction in the tensile strength and a slight increase in the elongation at break at 2.5% PLA.[24] Remarkably, only in one study the materials were sufficiently dried prior to processing, but still far-reaching conclusions were drawn on the influence of PLA on the mechanical properties of rPET. None of these empirical studies processed rPET spiked with PLA in a manner that is normal to produce rPET bottles, namely, with sufficient drying of the materials, twin screw extrusion, drying, injection moulding and solid-state processing (SSP)[25,26] treatment. SSP treatment involves elevated temperatures at reduced pressures at which volatile contaminants are removed and the chain length of condensation polymers such as PET are restored.

In relation to the influence of PLA on the optical properties of rPET, there is only one empirical study and an outspoken design-guideline. Thoden van Velzen et al. reported in 2016 that the addition of 0.3% PLA to rPET resulting in pellets with the same optical properties as normal rPET after the full bottle recycling process (drying, twin screw extrusion, drying, injection moulding and SSP).[27] Conversely, a Quebecois design guideline claims that recycled PET containing 0.3% PLA is hazy and yellowish.[28]

Therefore two research questions emerge. First of all, what is the likelihood that PLA will be present in recycled PET that is used for blow moulding bottles in relation to the collection and sorting infrastructure present. And secondly, what is the effect of PLA on recycled PET after the complete processing steps are executed, including SSP and injection moulding. Therefore, the objective of this study is twofold. First, it aims to study the likelihood that PLA will be present in recycled PET and at what concentrations. Second, it aims to study the influence of the presence of PLA on the quality of recycled PET in realistic amounts, and to compare those effects to reference impurities that are currently present in rPET, namely, poly vinyl chloride (PVC) and poly (ethylene vinyl alcohol) (EVOH). This study is limited to the optical, thermal properties and the intrinsic viscosity.

The likelihood that an impurity is present in a recycled plastic can best be studied with models based on material flow analysis[29] that describe the collection and recycling systems in three steps (collection, sorting and mechanical recycling). These models use the composition of collected material and/or the composition of the sorted products as input values and calculate the composition of the recycled plastics with known transition coefficients for the main material (PET) and the relevant impurities during sorting and recycling.[27,30] These models are useful to quantify at which concentrations in the collected material the impurity in the recycled plastic reaches a critical threshold value.

The effect of an impurity on the quality of rPET and its suitability for blow moulding bottles can best be studied by mimicking the whole mechanical recycling process, hence including the post-condensation and injection moulding steps. The post-condensation step can namely elicit chemical reactions such as trans-esterification and thermal rearrangements. The injection moulding step is thermally the most demanding step, which can promote hydrolysis reactions and thermally activated rearrangements.

2 | MATERIALS AND METHODS

2.1 | Modelling

The expected concentration of impurities in PET recycling feedstocks was calculated by multiplying the concentrations in the sorted products with the transfer coefficients of mechanical recycling, see Equation S1. Polymeric impurities in sorted products are present in two forms: non-targeted plastic objects (sorting mistakes) and components of targeted packages. PLA[21] and PVC[31,32] as impurities originate from sorting mistakes and EVOH originates mostly from components of targeted packages (lidding film from PET trays, etc.).[27] Three recycling value chains were analysed: (1) the current deposit-refund system for PET bottles, (2) the separate collection of LWP resulting in a recycled PET made from predominantly bottles and (3) the separate collection of LWP resulting in a recycled PET made from predominantly trays.

2.2 | Materials

Transparent PET bottles (1.5 L) were obtained from a local, Dutch beverage company and were produced to contain carbonated soda drinks. The bottles were supplied without caps, labels, glue, and so forth, and were produced to qualify both food safety regulations and the design guidelines of the Dutch deposit refund system named SRN. Transparent PLA salad trays (360 ml) were purchased from Pacovis. Virgin pellets of EVOH (type EVAL F171B) was obtained from Kuraray Co. PVC was obtained as a flexible package that was retrieved from Dutch LWP, it was a package used for textiles by a local warehouse.

2.3 | Processing

Both the PET bottles and the PLA trays were first grinded separately into 1–3 cm flakes using a WEIMA WLK-04 mill with a 35-mm sieve plate. These flakes were subsequently cut into smaller flakes using a Wanner compact granulator C 17.26sv provided with a 6-mm sieve plate. Both comminutors were thoroughly cleaned before and after milling. The EVOH pellets were cryogenically milled to a fine powder. The PVC flexible package was cut in small flakes with scissors and subsequently cryogenically milled to a fine powder.

Prior to processing, materials were dried in a desiccant dryer. The PET flakes were dried for 8 h at 120°C and the PLA flakes for 8 h at 80°C. Also the EVOH and PVC powders were separately dried for 8 h at 80°C. After drying and prior to processing, the water content of PET was checked using Karl Fisher titration. Karl Fisher titrations were performed according to ISO 15512 B2 using a Mettler-Toledo DL38 Karl Fisher Coulometer. The temperature for vaporisation was 150°C. The targeted levels of water content were below 50 ppm for PET and below 150 ppm for PLA.

Compounds were prepared using a Berstorff ZE 40 A* 38 D co-rotating twin screw extruder provided with a standard mixing screw, melt filter and strand pelletiser. The throughput was 5 kg/h at 300 rpm. The temperature settings used during extrusion were the following; Zone 1 (feed hopper) at 100°C, Zone 2 at 200°C, Zone 3 at 240°C, Zones 4 to 9 at 260°C, melt-filter and die at 265°C. Grinded and dried materials were used as feedstock with the exception of the additives (PVC and EVOH) which were added in small amounts (0.1% and 0.3%) as powders to allow accurate dosing. The time in between sample collection was more than 20 min, to allow the extruder to flush clean. The pressure in the extruder before the filter was approximately 5 bar. The maximum temperature of melt was measured and ranged between 265°C and 270°C. The process was stable in all cases, only colour variations in the strands were observed.

The produced granulates were subjected to SSP to mimic the conventional PET recycling process. Prior to SSP treatment, the pellets were dried and crystallised in a drier at 120°C for 4 h. SSP treatments were performed at the Senbis institute (Emmen, the Netherlands) at 220°C for 8 h. A vacuum of 3 mbar was applied with constant nitrogen bleed during processing.

SSP-treated granulates were injection moulded into 50 × 50 × 2 mm plaques intended for haze and colour measurements. The test specimens were manufactured using a Sumitomo DEMAG IntElect75-250 injection moulding machine with a screw diameter of 18 mm. The maximum cylinder temperature was 270°C, the screw was speed 200 rpm and the mould temperature was 30°C. The pressures during injection moulding varied. For the 100% rPET sample, the pressure was approximately 1450 ± 50 bar The pressure was slightly lower for the sample with 1% PLA: 1360 ± 30 bar; for the sample with 0.1% PVC: 1250 ± 60 bar; and for the sample with 0.1% EVOH: 1230 ± 30 bar.

2.4 | Analysis

Colour measurements were performed using a Minolta Chroma Meter CR-400. The colour of plaques and bottles was measured against a white tile as background (the same white tile that is used to calibrate the colour measurement device). The colour of granulate was determined on a tray filled with 1-cm loose granulate. All measurements were performed in fivefold and averaged, yielding average CIElab values for L*, a* and b* and their standard deviation.[27] The same method was used to measure colour of pellets and plaques.

The intrinsic viscosity (IV) was measured according to ASTM D4603-03. There were 250-mg samples (±1%) dissolved in 50-ml solvent, a 60:40 mixture of phenol and tetrachloroethane at a temperature of 100°C during 30 min. The IV of the samples and solvent reference was measured at 30°C using a calibrated Cannon-Fenske capillary type 100. A Schott Geräte CT1250 thermostat water bath provided with a CK100 water cooler was used to maintain a constant temperature. The automated measurements were performed using a Schott Geräte AVS 350. The IV-values were calculated using the Billmeyer equation. Duplicate measurements were performed on individually prepared samples. Averages and the standard deviation were reported.

The haze was measured at Holland Colours (Apeldoorn) according to ASTM D1003 using a Byk HazeGuard apparatus for transmission, haze and clarity. The haze was measured on bottle walls and injection moulded plaques. Measurements were performed in fivefold, and averages are reported including the standard deviation.

Differential scanning calorimetry (DSC) measurements were performed using a Perkin Elmer DSC 8000 provided with liquid nitrogen cooling and an auto sampler. Stainless steel DSC cups with rubber ring were used. The temperature programme was composed of the first heating from 40°C to 280°C, a cooling run to 40°C and a second heating run to 280°C, all at 10°C/min. The measurements were performed on the pellets that were compounded and crystallised, on the pellets that were subsequently treated with SSP and on the injection moulded plaques. The degree of crystallinity was calculated from the melting enthalpy in the second heating run, according to the method of Torres et al.[33]

3 | RESULTS

3.1 | Modelled concentration of impurities in recycled PET

The modelled concentration of PLA in three types of recycled PET from the current value chains are listed in Table 1, the data used and its sources are listed in Table S1.1, Table S1.2 and Table S1.3. These expected concentrations of PLA as impurity in PET vary with the collection system; ranging from absolute 0% for the deposit refund system to 0.019% in the recycled PET product made from PET trays originating from LWP collection. These estimations are based on measurements of sorted products when there were still few PLA packages on the Dutch markets and hence, these numbers will rise when more PLA based packages are introduced.

TABLE 1 The expected current concentration of PLA as an impurity in recycled PET pellets for three recycling value chains

Recycling value chain	Expected concentration of PLA in recycled PET pellets, [%]
1. DRS, PET bottles	0.000%
2. LWP, PET bottles	0.011%
3. LWP, PET trays	0.019%

Alearts et al. report higher expected concentrations for PLA in recycled PET products.[19] Their numbers are based on expected Belgian sales data and not from actual compositions of recycled PET products or sorted PET products. Nevertheless, we agree that concentrations can rise when their market share grows.

Bergsma et al. estimated that the current concentration of PLA in separately collected Dutch LWP is roughly 0.1%–0.4%. They suggest that this concentration could rise to 4%–5% in 2030.[15] Empirical sorting tests at these higher concentrations of PLA in LWP resulted in sorted PET products with PLA concentrations of 0.02% in case the PLA sorting machine was placed first in the NIR cascade of the sorting facility. However, when the PLA sorting machine was placed after the NIR sorting machine for PET, the PLA concentration in the PET sorted product rose to 0.91%–0.96%.[15] Due to the mechanical recycling process which commonly involves flake sorters for these materials, the concentration of PLA in the recycled PET pellets will typically be a factor 10 lower,[27] so roughly 0.1%. These results show that with the right technical precautionary measures the PLA concentration in PET sorted products can be controlled to sub-percent levels, even if the consumption of PLA increases largely. The concomitant downside of purifying PET with flake sorting machines is, however, that it will result in lower PET yields.

3.2 | Effect of impurities on the quality of rPET

3.2.1 | Colour of the pellets

The CIElab colour values (L*, a*, b*) of the pellets before crystallisation, after crystallisation and after SSP treatment are listed in Table S2.1. Furthermore, a picture is shown in Figure 1 of the pellets after SSP treatment. All pellets are greyish. The pellets with 0.1% PVC are clearly darker coloured as is eminent from the picture and from the lower L* and higher a* and b* values. The pellets with PLA

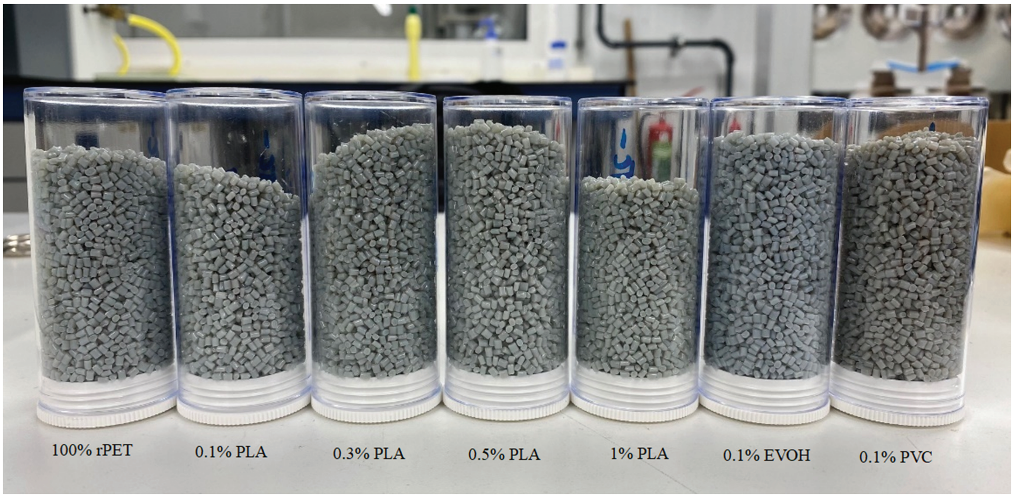


FIGURE 1 Photo of the pellets after SSP treatment

tend to be a little bit more yellow and green, as is apparent from the slightly lower a* and the slightly higher b* values at the higher levels of PLA. Pellets with EVOH have slightly lower a* and b* values.

During the processing steps, the L* values tend to rise, meaning that the pellets become lighter coloured. The a* values drop during the SSP treatment, which implies that the pellets become greener. The b* values tend to be lower for pellets after the SSP treatment in comparison to pellets before crystallisation.

3.2.2 | Colour values and haze of the plaques

The CIElab colour values of the 2-mm thick plaques are listed in Table 2, a photo is shown in Figure 2 and the relation between impurity concentration and CIElab values is shown in Figure 3. The L* value increases slightly with PLA as impurity, indicating that the recycled PET with PLA becomes more light and transparent. Conversely with only 0.1% PVC the L* values reduces substantially, implying that this recycled PET has become darker coloured. The a* value reduces slightly with increasing PLA concentrations, implying that these samples of recycled PET coloured slightly greenish. The b* value was found to increase slightly with level of PLA indicating slight yellowing. However, the sample with 0.1% of PVC had a two-fold higher b* values than the samples with PLA, indicating strong yellowing.

Three plaques (100% rPET, 1% PLA and 0.1% PVC) were annealed at 100°C for 1 min and the haze measurement was repeated to mimic the thermal exposure that preforms experience during reheating prior to bottle blowing. The haze values for the 100% rPET and 0.1% PVC were not influenced by the annealing step, whereas the haze value for the 1% PLA plaque was slightly raised with only one percent point, see Table S3.1. This could indicate that the PLA within the PET plaque crystallises during this treatment. In this respect, it is worth noting that the grade of PLA that was used, is relative rich in the D-isomer of lactic acid. This grade of PLA crystallises relatively slowly.

TABLE 2 The CIElab colour values and haze values of 2-mm thick plaques of rPET with impurities at various levels

	L*	a*	b*	Haze, [%]
100% rPET	65.7 ± 0.4	−1.6 ± 0.1	16.3 ± 0.2	19.4 ± 1.8
0.1% PLA	68.5 ± 0.1	−2.3 ± 0.1	14.6 ± 0.0	11.2 ± 0.3
0.3% PLA	68.0 ± 0.2	−2.6 ± 0.3	16.4 ± 0.0	11.2 ± 0.3
0.5% PLA	67.2 ± 0.3	−2.2 ± 0.1	16.7 ± 0.1	14.2 ± 0.1
1% PLA	67.2 ± 0.1	−2.6 ± 0.1	18.4 ± 0.1	13.2 ± 0.1
0.1% EVOH	65.4 ± 0.3	−1.5 ± 0.2	11.8 ± 0.1	12.7 ± 0.4
0.1% PVC	62.5 ± 0.3	−1.2 ± 0.1	27.3 ± 0.2	10.8 ± 0.4

3.2.3 | Intrinsic viscosity

The intrinsic viscosity values of the pellets have clearly decreased as compared to the initial value of the grinded bottles, see Table 3. This is common, as recycled PET contains small amounts of water that result in a limited hydrolysis during thermal processing and hence in lower IV values.[33,34] After the solid-state processing step, the intrinsic viscosities were raised, which indicates that the molecular weights have been restored. The impurities did have an influence on the final intrinsic viscosities, as they slowdown the restoration of the intrinsic viscosities during SSP. This is in agreement with previous observations.[27] This negative effect of an impurity was the largest for PVC. Only the rPET with 0.1% PVC did not attain the targeted level of 0.8 g/dl. The rPET sample with 0.1% EVOH exceeded the target level, but its IV was clearly reduced as compared to the 100% rPET sample. Of all the impurities tested, PLA was found to have the least effect on the restoration of the intrinsic viscosity. All the rPET samples with

FIGURE 2 Photo of the 5 × 5 × 0.2 cm plaques of the recycled PET with various impurities, clearly showing the colour

PLA as impurities reached the target IV value, and no clear relation was found between IV value reached and the concentration of PLA impurity in the rPET.

3.2.4 | Thermal properties

The deliberately added impurities were found to have very limited effects on the thermal properties as determined with DSC. This was found for the three types of samples that were tested: (1) the rPET pellets after compounding and crystallisation but before SSP treatment; (2) the rPET pellets after compounding, crystallisation and SSP treatment; and (3) the plaques (that have been subjected to the whole process including SSP and injection moulding). The results are listed in Table S4.1 for the crystallised pellets before SSP treatment, in Table S4.2 for the pellets after SSP treatment and Table S4.3 for the injection moulded plaques. The degrees of crystallinity of the samples are listed in Table S4.4.

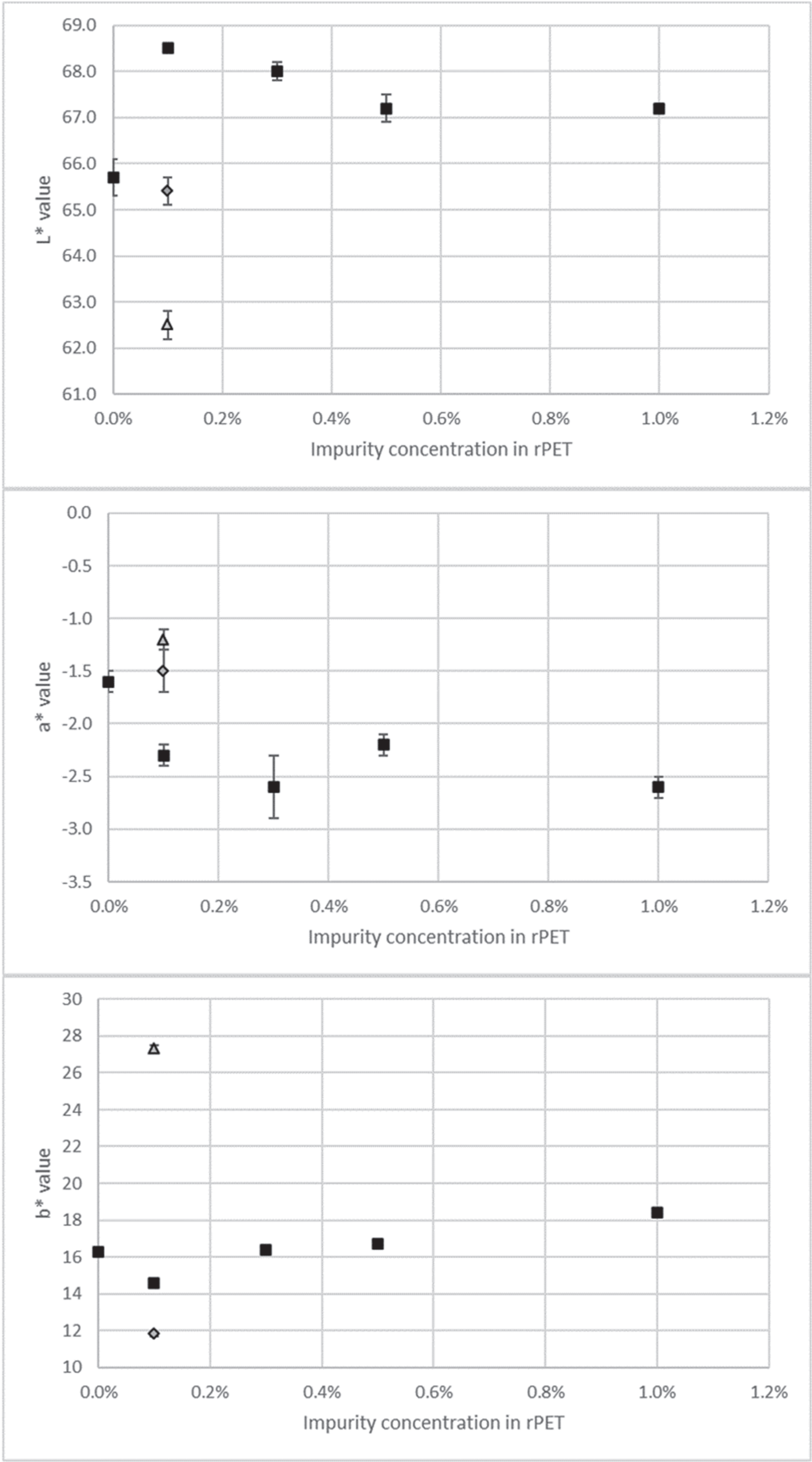
The SSP treatment raised the melting enthalpy and the degree of crystallisation for all samples. One of the small noticeable differences was the difference in onset melting temperature between the grinded bottle material and the rPET pellets. For the grinded PET bottle material this onset melting temperature was 238.8°C, whereas it was 243.3°C for the compounded and crystallised pellets, see Table S4.1. A likely explanation is that the grinded PET material contains a fraction of PET chains with short chain lengths, which is better integrated in the crystallites of the crystallised pellets. Another small noticeable difference was found in the degree of crystallinity of the pellets after SSP treatment. The pellets containing PLA had slightly lower degrees of crystallinity as compared to the other samples (100% rPET, 0.1% PVC and 0.1% EVOH). But after injection moulding, the degrees of crystallisation for most samples were comparable again, with the exception of the sample with 0.1% PVC, which clearly had a raised degree of crystallinity. This indicates that this sample with PVC has lower molecular weights.

4 | DISCUSSION

4.1 | Impact of the recycling method on the optical properties

The optical properties of all the produced rPET plaques, including the 100% rPET plaque, exceed industrial acceptance limits. The Association of Plastic Recyclers defined the limits for the optical properties of 3-mm thick PET plaques to be: L* has to exceed 84, b* value should not exceed 4 and the haze value should be below 9.5%.[35] Since there are no methods to correct these optical properties for thickness effects, it is simply concluded that the samples made in the laboratory are less transparent and more coloured than in the industrial practice. The exceedance is most prominent for the transparency (L* and haze) and the yellowness (b*). Since also the 100% rPET plaque has optical properties for 2-mm plaques that exceeds the specifications for 3-mm plaques, it is concluded that the chosen method causes the samples to be systematically, insufficiently transparent and too yellow. This has been reported previously for laboratory processes that mimic the industrial processing of rPET[36] and is likely to be caused by the smaller scale, which results in more contact area and longer exposure times to elevated temperatures. Therefore, the results of this laboratory study should not be used absolutely, but only in comparative manner to study the relative impact of contaminants on the optical properties.

FIGURE 3 The relations between the CIElab colour values of the PET plaques in relation to the concentration of impurities. ■ = PLA, ♦ = EVOH and ▲ = PVC



4.2 | Influence of impurities on recycled poly (ethylene terephthalate)

This study has reconfirmed that PVC has a significant detrimental impact on the optical properties of rPET, the intrinsic viscosity and the degree of crystallinity. Already at a concentration of 0.1% PVC, the rPET becomes more yellow, grey and hazy. In previous research that has been explained,[36] during the thermal conversion of the rPET, the PVC impurity decomposes forming hydrochloric acid, which catalyses rearrangements in the PET polymeric backbone forming quinone-like structures that are yellow. Additionally, PVC and decomposed PVC both do not mix with PET and form a blend morphology, which will increase the haziness. Finally, the presence of hydrochloric acid in the rPET causes the backbone to hydrolyse and

TABLE 3 Intrinsic viscosity values [g/dl] of the grinded PET bottle feedstock and the recycled PET pellets produced with various levels of impurities

	IV after crystallisation	IV after SSP
Grinded PET bottles	0.77 ± 0.01	na
100% rPET	0.66 ± 0.02	1.05 ± 0.02
0.1% PLA	0.62 ± 0.01	0.89 ± 0.06
0.3% PLA	0.65 ± 0.02	0.96 ± 0.02
0.5% PLA	0.66 ± 0.01	0.97 ± 0.09
1% PLA	0.65 ± 0.01	0.93 ± 0.01
0.1% EVOH	0.64 ± 0.02	0.86 ± 0.03
0.1% PVC	0.60 ± 0.02	0.79 ± 0.03

Abbreviation: na, not applicable.

counteracts the SSP treatment in restoring the chain lengths, which become evident as a relatively low intrinsic viscosity and a raised degree of crystallinity.

This study did not find an effect of the presence of 0.1% EVOH on the optical and thermal properties of rPET. Perhaps EVOH is miscible with PET at these low levels or it is partially trans-esterified with PET, as previously suggested,[27] but that cannot be corroborated or refuted from these results. The only negative impact of the presence of 0.1% EVOH was that it apparently retards the recondensation of PET during the SSP treatment processing, as is evident from the lower IV value after SSP treatment than the normal rPET.

Similarly, this study did not reveal any negative impacts of the presence of 0.1% to 1% of PLA as an impurity in PET on the optical and thermal properties. Also the intrinsic viscosity is not affected. Since PLA-containing rPET is not more hazy than normal rPET, PLA appears not to form a blend structure in PET at the studied concentration range (0.1%–1%). This is in agreement with previous research[24] that have report partial miscibility at concentrations below 10%.

The remaining question is why did not our study find a clear impact of PLA on the optical properties of rPET, whereas other studies have reported those impacts. The most likely reason for the contradictory results is the difference in processing. This study deliberately processed rPET in the demanding manner that is common in rPET production of food grade rPET for bottles. Hence, we mixed with a double screw extruder, dried the material, subjected the material to SSP and injection moulded it. In other studies, a kneader or a single screw extruder was used and the material was not dried or treated with SSP. Apparently, this more light processing method creates a different morphology for the mixture with different optical properties. This observation reconfirms the importance of proper processing methods in recycling studies.

To avoid a potential negative impact of PLA on the quality of rPET in the future we should aim to maintain the PLA concentration below 1% in rPET pellets. Since, the sorted PET product trays (DKR 328-5) is the most susceptible to the inclusion of PLA trays, we have calculated what the maximum concentration of PLA in the sorted product PET trays should be with Equation S1. By taking the masses and transfer coefficient listed in Table S1.3, this concentration limit for the sorted product is 8%. Due to the fact that PLA objects have distinctively different NIR spectra than objects made from PET,[13] these types of limits are easy to accomplish in modern sorting facilities.

4.3 | Implications for the circular economy

This study has shown that, currently, PLA is hardly present in rPET originating from the Netherlands. The concentration varies from absolute zero for rPET made from DRS bottles to maximally 0.019% for rPET made from PET trays from LWP collection. Furthermore, when the consumption of PLA packages will rise in the future, this concentration of PLA in rPET will hardly rise, since most PLA objects will be sorted out with regular NIR sorting machines in a modern sorting facility[15] and most of the PLA that has slipped through will subsequently be removed with NIR flake sorting machines, which are common in PET recycling facilities that process sorted PET products that originate from LWP collection. Even in case that the concentration of PLA trays would rise to 8% in sorted PET product, the PLA concentration in the rPET will remain below 1%. Additionally, when 0.1% to 1% of PLA was added to rPET and this mixture was thoroughly extruded, dried, subjected to SSP and injection moulded, the optical and thermal properties were not affected. This suggests that the use of PLA in packaging applications does not endanger the recycling of PET bottles and trays. Therefore, new biobased polymers can be integrated in existing LWP management infrastructures with modern NIR-based sorting machines. In order to make this attractive for the incumbents, a new sorting category such as PLA or biobased polyesters should be created and sufficient recycling infrastructure should be erected to process these polyesters. This will enable the decoupling of fossil based feedstocks for the plastic system[6] and gradually also enable a transition towards intrinsic sustainable packages.[37] The latter requires the plastic system not only to decouple from fossil feedstocks and to minimise environmental impacts, but also to render the plastic objects bio-compatible (hence avoiding damage when plastics leak into nature).

5 | CONCLUSION

The effect of PLA on PET recycling has been studied. The optical and thermal properties of PET were not found to be affected by the presence of 0.1% to 1% of PLA. The current concentration of PLA in Dutch recycled PET was estimated by modelling to be between absolute zero and 0.019%. Therefore, at these current low concentrations PLA will not jeopardise the recycling of PET. Even if the consumption of PLA packages would rise largely in the future, then the concentration of PLA in rPET can be maintained below 1% by a combination of rigorous NIR sorting in the sorting facility and flake sorting in the recycling facility. Therefore, this study suggests that it is possible to gradually integrate biobased plastics with the current LWP management system, which is necessary to reduce the environmental impacts of the plastic system.

ACKNOWLEDGEMENTS

We acknowledge Herman de Beukelaer for his help with the DSC measurements. Maarten Ubbink of Holland Colours Apeldoorn is acknowledged for performing the haze measurements. Marieke T. Brouwer and Maelenn Ravard are thanked for their meticulous review and suggestions.

CONFLICT OF INTEREST

The first three authors (Eggo Ulphard Thoden van Velzen, Sharon Chu and Karin Molenveld) report no conflict of interest. Vladislav Jašo, the fourth author, works in the R&D Department of Total Corbion PLA.

FUNDING INFORMATION

This study was funded by Total Corbion PLA.

DATA AVAILABILITY STATEMENT

The data that supports the findings of this study are available in the supplementary material of this article.

ORCID

Eggo Ulphard Thoden van Velzen https://orcid.org/0000-0002-2174-3220

REFERENCES

1. European Commission. *A European Strategy for Plastics in a Circular Economy*. Brussels: European Commission; 2018:16.
2. Crippa M, de Wilde B, Koopmans R, et al. In: De Smet M, Linder M, eds. *A Circular Economy for Plastics—Insights from Research and Innovation to Inform Policy and Funding Decisions*. Brussels, Belgium: European Commission; 2019:244.
3. Brouwer MT, Thoden van Velzen EU, Ragaert K, ten Klooster R. Technical Limits in Circularity for Plastic Packages. *Sustain For*. 2020; 12(23):10021 doi:10.3390/su122310021
4. Hahladakis JN, Iacovidou E. An overview of the challenges and trade-offs in closing the loop of post-consumer plastic waste (PCPW): focus on recycling. *J Hazard Mater*. 2019;380:120887. doi:10.1016/j.jhazmat.2019.120887
5. Gerassimidou S, Martin OV, Chapman SP, Hahladakis JN, Iacovidou E. Development of an integrated sustainability matrix to depict challenges and trade-offs of introducing bio-based plastics in the food packaging value chain. *J Clean Prod*. 2021;286:125378. doi:10.1016/j.jclepro.2020.125378
6. World Economic Forum EMF. *The New Plastics Economy—Rethinking the Future of Plastics: Ellen MacArthur Foundation*; 2016:1-120.
7. Cimpan C, Bjelle EL, Strømman AH. Plastic packaging flows in Europe: a hybrid input–output approach. *J Ind Ecol*. 2021;25(6):1-16. doi:10.1111/jiec.13175
8. Kral U, Morf LS, Vyzinkarova D, Brunner PH. Cycles and sinks: two key elements of a circular economy. *J Mat Cycles Waste Manag*. 2018; 21(1):1-9. doi:10.1007/s10163-018-0786-6
9. Hahladakis JN, Iacovidou E. Closing the loop on plastic packaging materials: what is quality and how does it affect their circularity? *Sci Total Environ*. 2018;630:1394-1400. doi:10.1016/j.scitotenv.2018.02.330
10. Madival S, Auras R, Singh SP, Narayan R. Assessment of the environmental profile of PLA, PET and PS clamshell containers using LCA methodology. *J Clean Prod*. 2009;17(13):1183-1194. doi:10.1016/j.jclepro.2009.03.015
11. Maga D, Hiebel M, Thonemann N. Life cycle assessment of recycling options for polylactic acid. *Resour Conserv Recycl*. 2019;149:86-96. doi:10.1016/j.resconrec.2019.05.018
12. Bishop G, Styles D, Lens PNL. Environmental performance comparison of bioplastics and petrochemical plastics: a review of life cycle assessment (LCA) methodological decisions. *Resour Conserv Recycl*. 2021;168:105451. doi:10.1016/j.resconrec.2021.105451
13. Desole MP, Aversa C, Barletta M, Gisario A, Vosooghnia A. Life cycle assessment (LCA) of PET and PLA bottles for the packaging of fresh pasteurised milk: the role of the manufacturing process and the disposal scenario. *Packag Technol Sci*. 2021;1-18. doi:10.1002/pts.2615
14. Chen X, Kroell N, Li K, Feil A, Pretz T. Influences of bioplastic polylactic acid on near-infrared-based sorting of conventional plastic. *Waste Manag Res*. 2021;39(9):1210-1213. doi:10.1177/0734242X211003969
15. Beeftink M, Vendrik J, Bergsma G, van der Veen R. *PLA Sorting for Recycling, Experiments Performed at the National Test Centre of Circular Plastics*. CE Delft; 2021:21.190180.048.
16. Soroudi A, Jakubowicz I. Recycling of bioplastics, their blends and biocomposites: A review. *Eur Polym J*. 2013;49(10):2839-2858. doi:10.1016/j.eurpolymj.2013.07.025
17. Badia JD, Ribes-Greus A. Mechanical recycling of polylactide, upgrading trends and combination of valorization techniques. *Eur Polym J*. 2016;84:22-39. doi:10.1016/j.eurpolymj.2016.09.005
18. Beltran FR, Barrio I, Lorenzo V, Del Rio B, Martinez Urreaga J, de la Orden MU. Valorization of poly (lactic acid) wastes via mechanical recycling: improvement of the properties of the recycled polymer. *Waste Manag Res*. 2019;37(2):135-141. doi:10.1177/0734242X18798448
19. Alaerts L, Augustinus M, Van Acker K. Impact of bio-based plastics on current recycling of plastics. *Sustain For*. 2018;10(5):1-15. doi:10.3390/su10051487
20. Wojnowska-Baryła I, Kulikowska D, Bernat K. Effect of bio-based products on waste management. *Sustain For*. 2020;12(5):1-12. doi:10.3390/su12052088
21. La Mantia FP, Botta L, Morreale M, Scaffaro R. Effect of small amounts of poly (lactic acid) on the recycling of poly (ethylene terephthalate) bottles. *Polym Degrad Stab*. 2011;97:21-24. doi:10.1016/j.polymdegradstab.2011.10.017
22. McLauchlin AR, Ghita OR. Studies on the thermal and mechanical behavior of PLA-PET blends. *J Appl Polym Sci*. 2016;133(43):1-11. doi:10.1002/app.44147
23. Gere D, Czigany T. Future trends of plastic bottle recycling: compatibilization of PET and PLA. *Polym Test*. 2020;81:106160. doi:10.1016/j.polymertesting.2019.106160
24. Aldas M, Pavon C, De La Rosa-Ramírez H, et al. The impact of biodegradable plastics in the properties of recycled polyethylene terephthalate. *J Polym Environ*. 2021;29(8):2686-2700. doi:10.1007/s10924-021-02073-x
25. Welle F. Twenty years of PET bottle to bottle recycling—an overview. *Resour Conserv Recycl*. 2011;55(11):865-875. doi:10.1016/j.resconrec.2011.04.009
26. Fitaroni LB, de Oliveira EC, Marcomini AL, Paranhos CM, Freitas FL, Cruz SA. Reprocessing and solid state polymerization on contaminated post-consumer PET: thermal and crystallization behavior. *J Polym Environ*. 2020;28(1):91-99. doi:10.1007/s10924-019-01579-9
27. Thoden van Velzen EU, Brouwer MT, Molenveld K. *Technical Quality rPET: Wageningen Food & Biobased Research, Wageningen (NL)*. 2016; 1661:147. https://edepot.wur.nl/392306

28. Quebec EE. Factsheet 3 impact of packaging on curbside recycling collection and recycling system: PLA bottle. In: *Secondary Factsheet 3 impact of packaging on curbside recycling collection and recycling system: PLA bottle April 25th 2012*; 2012. http://www.eeq.ca/wp-content/uploads/PLA-bottles.pdf
29. Brunner PH, Rechberger H. *Handbook of Material Flow Analysis*. Boca Raton FL: CRC Press; 2020.
30. Brouwer M, Picuno C, Thoden van Velzen EU, Kuchta K, De Meester S, Ragaert K. The impact of collection portfolio expansion on key performance indicators of the Dutch recycling system for post-consumer plastic packaging waste, a comparison between 2014 and 2017. *Waste Manag*. 2019;100:112-121. doi:10.1016/j.wasman.2019.09.012
31. Paci M, La Mantia FP. Influence of small amounts of polyvinylchloride on the recycling of poly ethylene terephthalate. *Polym Degrad Stab*. 1999;63(1):11-14. doi:10.1016/S0141-3910(98)00053-6
32. Pawlak A, Pluta M, Morawiec J, Galeski A, Pracella M. Characterization of scrap poly (ethylene terephthalate). *Eur Polym J*. 2000;36(9):1875-1884. doi:10.1016/S0014-3057(99)00261-X
33. Torres N, Robin JJ, Boutevin B. Study of thermal and mechanical properties of virgin and recycled poly (ethylene terephthalate) before and after injection molding. *Eur Polym J*. 2000;36(10):2075-2080. doi:10.1016/S0014-3057(99)00301-8
34. Awaja F, Pavel D. Recycling of PET. *Eur Polym J*. 2005;41(7):1453-1477. doi:10.1016/j.eurpolymj.2005.02.005
35. APR. *PET Heat History and Discoloration Evaluation. Secondary PET Heat History and Discoloration Evaluation November 16th 2018*. 2018. https://plasticsrecycling.org/images/Design-Guidance-Tests/APR-PET-S-03-heat-history-discolor-screening.pdf
36. Alvarado Chacon F, Brouwer MT, Thoden van Velzen EU. Effect of recycled content and rPET quality on the properties of PET bottles, part I: optical and mechanical properties. *Packag Technol Sci*. 2020;33(9):1-11. doi:10.1002/pts.2490
37. Packaging NIfS. *The State of Sustainable Packaging*. The Hague: Netherlands Institute for Sustainable Packaging; 2020. https://kidv.nl/media/rapportages/kidv-publication-sosp-en-a4-pdf_def.pdf

SUPPORTING INFORMATION

Additional supporting information may be found in the online version of the article at the publisher's website.

How to cite this article: Thoden van Velzen EU, Chu S, Molenveld K, Jašo V. Effect of poly lactic acid trays on the optical and thermal properties of recycled poly (ethylene terephthalate). *Packag Technol Sci*. 2022;35(4):351-360. doi:10.1002/pts.2633