RESEARCH ARTICLE

Strategies for improving product properties of thermoformed multilayer cups with contents of post-consumer polypropylene recyclates

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Abstract

Thermoformed products bear great potential for the application of recycled materials when using multilayer structures for incorporating post-consumer recyclates. For this study, four different commercially available polypropylene recyclates are selected as possible core layers for the processing of multilayer thermoformed products with top layers of virgin polypropylene. Tests are performed on material level as well as on product level. At the material level, recyclates exhibit different mechanical property profiles with lower stiffness but higher toughness values due to polyethylene contamination. At the product level, thermoformed cups with core layers of recyclate material show lower shrinkage than cups made from polypropylene virgin material only. Most cups with recycled content achieve lower top load than the control cup. Based on these results, three strategies for improving product performance are defined and tested. Namely, (i) the use of a higher share of top layers, (ii) the blending of the recyclate core layer with a polypropylene virgin material with higher stiffness, and (iii) the increase in overall film thickness. All the strategies to adjust the film structure achieve better results in terms of shrinkage behavior and top load. However, the strategy that focuses on increasing the overall film thickness is not economically feasible.

KEYWORDS

characterization, design from recycling, mechanical properties, plastics recycling, polypropylene, post-consumer recyclate, strategies

1 | INTRODUCTION

A wide range of stakeholders from industry, academia, legislation and consumers is pushing for a more responsible approach to the production, consumption, and disposal of plastics.[1–4] Although it has long been common practice to recycle and reuse packaging made from glass, metal and paper, recycling of plastics packaging is still lagging. Roughly, 10% of the plastics used for products are recycled plastics.[5] Post-industrial recycling of production waste, such as edge trims, discarded parts, and punching grids is already being carried out very widely for economic reasons, as this usually uncontaminated waste can be returned to the production cycle

without intense pretreatment effort. The recycling of post-consumer waste is much more difficult, which is why the European Union, for example, is assisting the industry with guidelines. The current directive 2018/852 stipulates that 50% of plastics packaging waste must be recycled by the end of 2025 and 55% by the end of 2030.[6] Strategies known as "design for recycling" and "design from recycling" provide a guideline for the use of recycled materials. Thus, products need a design which allows for recycling (e.g., avoiding a mix of materials, easy separation) and a design which allows the uptake of recyclates.[7]

Although mechanically recycled post-consumer polyolefin recyclates are in high demand on the market, they cannot compete with virgin polyolefin in terms of material properties. While it is common practice to separate low-density and high-density polyethylene (PE), in the case of polypropylene (PP), different types (homopolymers and copolymers), and grades designed for different processing methods are mixed in the recycling process, as they are usually not collected separately. On the one hand, modern sorting plants are not yet capable of achieving this sorting depth and, on the other hand, recycling companies do not even strive for this for economic reasons. In most cases, the melt flow rate (MFR) of commercially available post-consumer PP recyclates (rPPs) starts at 10 g/10 min.[8] Although PP grades with this MFR range are readily usable for processing by injection molding, they are less suitable for extrusion and thermoforming since a certain melt stability is required. The mechanical properties of rPPs are in the range of copolymers, which makes it again difficult to use for thermoforming applications that usually require high stiffness.[9] The impact behavior of rPPs is strongly influenced by macroscopic inclusions due to inorganic/organic residues in the waste fractions.[10] Furthermore, microscopic particles, additive residues and volatiles were found in further work.[11–14] These contaminations often lead to unpleasant odors and severely restrict the use of recyclates for food packaging. Furthermore, recyclates are often only offered in shades of gray or green, since the pretreated plastics waste is usually not additionally sorted by color.[14,15]

Recently, the share of products with recycled plastics content has increased significantly. A large part of this is recycled polyethylene terephthalate (rPET), as there are already some recycling processes for this material that are approved to produce rPET for food contact applications.[16] For PET bottles, there are well-implemented closed loops in the form of deposit systems. This ensures a highly clean material fraction. However, this type of waste collection only works for closed containers that are countable.[17] Polyolefin recyclates are currently often used for low-quality products. Some examples are listed in the literature.[18–20] However, the trend is toward the use of recycled polyolefin material in packaging for cleaning agents and hygiene products (rinse-off cosmetics), as the requirements for the material used for these applications are significantly lower than for food packaging.[21]

A study has shown that the use of multilayer structures of extruded PE films with encapsulated recyclate layer greatly reduces migration, which would make food contact suitability conceivable.[22] The investigation of injection molded PP multilayer structures in transport boxes revealed insufficient mechanical properties, which could not be compensated by cover layers of virgin PP and thus the material behavior of the recyclate always dominated.[10] However, multilayer structures could be a valuable tool to combine properties of several materials.[10,22–25] PE and PP recyclates can be modified in such a way by producing compounds with virgin material that they fulfill the requirements for blow molding and thermoforming. The material properties in short-term tests showed properties as to be expected in virgin material, but these materials showed clear deficits in the long-term tests such as environmental stress cracking resistance (ESCR). However, by mixing PP recyclates with virgin PP, the required property window was met for certain mixing ratios.[26,27]

The objective of this work is to evaluate the applicability of certain strategies to improve product properties of thermoformed cups, which fulfill the design from recycling approach and are thus partly made with recyclate. Four commercially available post-consumer PP recyclates were characterized at material level regarding their thermal and mechanical properties and were compared to standard virgin PP for packaging applications to determine the applicability for thermoforming applications. Therefore, differential scanning calorimetry (DSC) and thermogravimetric (TGA) measurements were performed to gain information about the material composition. Mechanical properties were determined using tensile test and Charpy impact test. Symmetric multilayer films with virgin PP top layers and PP recyclate core layers were extruded and subsequently processed to thermoformed cups. Three cup design strategies were defined, and the resulting thermoformed cups were tested regarding their quality control properties. These essential product properties are shrinkage diameter and top load. The first strategy comprised an increase in the thickness of the top layers and an associated decrease in the thickness of the core layer and thus of the absolute recyclate content. Blending a certain amount of virgin PP to the rPP for adjusting the core layer was defined as the second strategy. As a result of the blending, the flowability of the core layer material was decreased to reduce the probability of flow instabilities.[28] Both strategies were made while

maintaining the same overall thickness of the thermoforming film. The third strategy was to increase the overall thickness of the thermoforming film although the amount of material required increases when using this strategy. Nevertheless, the relative recyclate content remained constant in the third strategy.

2 | MATERIALS AND METHODS

2.1 | Materials

Two virgin PP grades and four different commercially available post-consumer PP grades were used for the following evaluations. The virgin PP homopolymers PP1 and PP2, one tailored for thermoforming and one for injection molding, respectively, were used as reference thermoforming material (PP1) and for varying multilayer structures (PP1 and PP2) with MFR values of 4 and 12 g/10 min, respectively. For thermoforming processes, materials with a high melt stability are needed as the films are heated up using infrared heaters and furthermore the hot films are drawn down. The relevant material behavior for this corresponds to a high molar mass, which affects the MFR to lower values.[29] The four different post-consumer PP grades rPP1, rPP2, rPP3, and rPP4 were supplied from three different companies with MFR values of 16, 16, 6, and 14 g/10 min, respectively. Most of these MFR values are rather high for thermoforming processes. These recyclates were the best we found in terms of their MFR and their commercial availability in high amounts to enable the use for industrial processing which requires at least a few 100 kg of granules. The availability of low MFR PP recyclates is scarce, as a lot of waste from injection molded parts is available and PP tends to increase the flowability during recycling due to chain scission.[30] Two of these recyclates are dark gray (rPP1 and rPP4), one is off-white (rPP2) and one is transparent with a slight brown discoloration (rPP3).

2.2 | Specimen preparation and material characterization

For DSC and TGA measurements, the granules were used as delivered. Multipurpose specimen type 1A and Charpy specimens were used for all mechanical testing methods.[31,32] They were injection molded with an Engel Victory 60 injection molding machine (Schwertberg, Austria) following ISO 294.[33] The specimens were stored at testing conditions for at least 96 h prior testing.

Thermoanalytical measurements to determine temperature points and enthalpies of melting events for the identification of different semi-crystalline polymers were carried out on a PerkinElmer DSC8500 (Waltham, MA). Aluminum pans were filled with samples of around 5 mg. The measurement included two heating steps in a range from 0 to 200°C and an intermediate cooling step with a heating and cooling rate of 10°C/min. Nitrogen was used as a purge gas for all scanning steps. All endothermic events of the second heating step were evaluated in J/g. Duplicates were measured per material.

Thermogravimetric analysis was utilized out to evaluate the residual amount of inorganic matter after pyrolysis. A PerkinElmer STA6000 (Waltham, MA) was used for the measurements. A sample of 40–60 mg was heated up from 30 to 800°C with a heating rate of 20°C/min in an inert nitrogen atmosphere. The residual mass at 700°C was evaluated. Two samples were tested per material.

Mechanical material parameters were determined via tensile test according to ISO 527-2 using a Zwick Roell Z005 universal testing machine (Ulm, Germany) equipped with MultiXtens II HP extensometers at ambient conditions of 23°C and a relative humidity of 50%.[32] The initial clamping length was set to 115 mm. According to the standard for the determination of the tensile modulus, the testing speed was 1 mm/min in a strain range from 0.05% to 0.25% and 50 mm/min for the rest of the measurement until rupture. Based on the measurement of five samples tensile modulus, yield stress and strain at break were evaluated.

The Charpy notched (NIS) and unnotched impact strengths (UIS) were determined to evaluate the ductility of the materials when they are subjected to an impact load. The tests were conducted using a Zwick Roell HIT25P impact tester (Ulm, Germany) according to ISO 179-1.[31] Therefore, 10 specimens per sample were notched on a Leica RM2265 microtome (Nussloch, Germany) with a Type A notch to obtain a remaining width of 8 mm. Additionally, 10 unnotched specimens were tested per material. Pendulums with 0.5, 2, and 15 J were used to stay in the proposed absorbed energy range of 10% to 80% of the pendulum as suggested in the standard. The specimens were tested edgewise at a temperature of 23°C and a relative humidity of 50%.

2.3 | Film setups, cup production, and product characterization

For the evaluation of the product quality, films and subsequently thermoformed cups were produced with a Kiefel KTR-4 thermoforming machine (Freilassing, Germany). First, a symmetric three-layer control cup was processed using PP1, the virgin PP with the lower MFR, as top layer and the in-house recyclate in the core layer. Moreover, multilayer cups for a screening of the different

recycled PP grades rPP1 to rPP4 were produced. Again, a symmetric three-layer structure (A-B-A) was used for all cups, where PP2, the virgin PP with the higher MFR, was used for the top layers. These setups are summarized in Table 1. Then, several cups were produced to evaluate the strategies (i), (ii), and (iii) as explained in Figure 1.

Based on the cup c1 the strategies (i) and (ii) were tested. Therefore, the cup c1_s with the same overall thickness but a different share (s) of the layers was produced. In this case, the top layers of cup c1_s are thinner than of cup c1. Hence, strategy (i) can be tested by comparing these cups. Subsequently, c1 and c1_s were modified using strategy (ii) by blending 15% virgin PP1 into the core layer. The lower MFR material was mainly used to reduce the probability of flow instabilities due to different MFR values of the top and core layer. The samples were named c1_b (b: blend) and c1_sb. Considering their recyclate content, these four cups can be ranked in the order c1_s > c1_sb > c1 > c1_b. This order will be used in the further evaluation. Regarding strategy (iii), the thickness of the initial thermoforming cup c2 was varied, where the film thickness was increased by up to 25% in three steps. The samples are named c2, c2_t8, c2_t17, and c2_t25, according to their thickness (t) increase in percent. The relative share of all layers and materials remained constant. A summary of all cup samples is depicted in Table 2.

At product level, shrinkage diameter, which represents the geometrical change due to the cooling of the upper open diameter of the cup, and top load tests were conducted. Thirty measurements on product level were done per material composition. For the shrinkage diameter, a certain predefined geometrical value was set to 100%. Furthermore, the 100% for the top load was defined by the value of the control cup made from virgin PP.

3 | RESULTS AND DISCUSSION

3.1 | Evaluation of material composition and properties at the material level

3.1.1 | Thermoanalytical properties

Thermoanalytical measurements are a practicable method to determine polymeric and non-polymeric components of a sample. Thermograms of PP1, PP2, and rPP1 to rPP4 are shown in Figure 2a. Vertical lines indicate the range of PE and PP melting events. The enthalpy of fusion of the PE fraction was determined from 115 to 132°C and of the PP fraction from 132 to 170°C. The absolute values of PE and PP melting points and enthalpies of fusion are given in Table 3. For comparison, melting points of polyethylene low density (PE-LD), linear low density (PE-LLD), medium density (PE-MD), and a polyethylene with high density (PE-HD) were

TABLE 1 Produced films/cups for a comparison of the selected recyclates.

Cup	Film setup	Comment
control	PP1	+ in-house recyclate
c1	PP2-rPP1-PP2	
c2	PP2-rPP2-PP2	
c3	PP2-rPP3-PP2	
c4	PP2-rPP4-PP2	
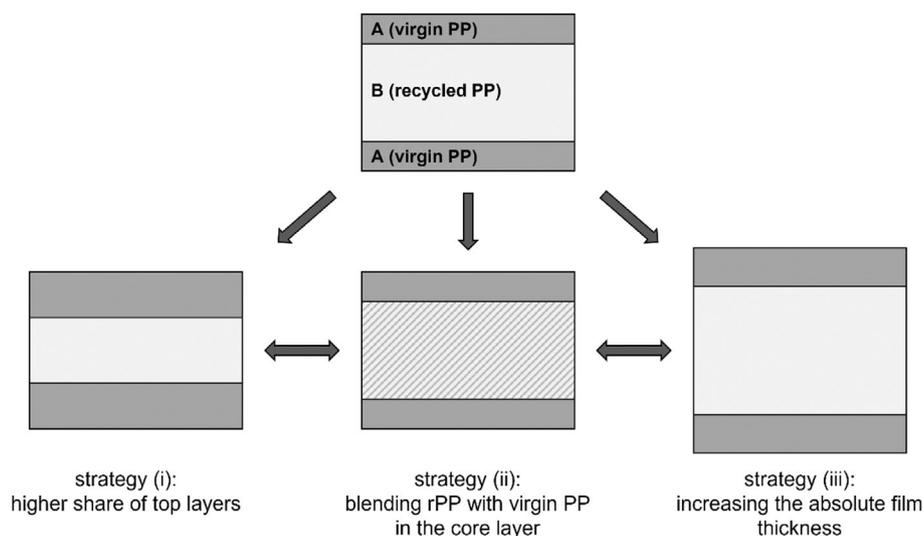
FIGURE 1 Graphical illustration of the tested modification strategies (i), (ii), and (iii).

TABLE 2 Produced films/cups for the evaluation of the three strategies, namely (i) higher share of top layers, (ii) blending PP recyclate with virgin PP for the core layer, and (iii) increasing the absolute film thickness.

Strategies (i) and (ii)			Strategy (iii)		
Cup	Film setup	Comment	Cup	Film setup	Comment
c1	PP2-rPP1-PP2		c2	PP2-rPP2-PP2	
c1_s	PP2-rPP1-PP2	Thinner top layer	c2_t8	PP2-rPP2-PP2	+ 8% thickness
c1_b	PP2-(rPP1 + PP1)-PP2	As film c1 + 15% PP1 in core layer	c2_t17	PP2-rPP2-PP2	+ 17% thickness
c1_sb	PP2-(rPP1 + PP1)-PP2	As film c1_s + 15% PP1 in core layer	c2_t25	PP2-rPP2-PP2	+ 25% thickness
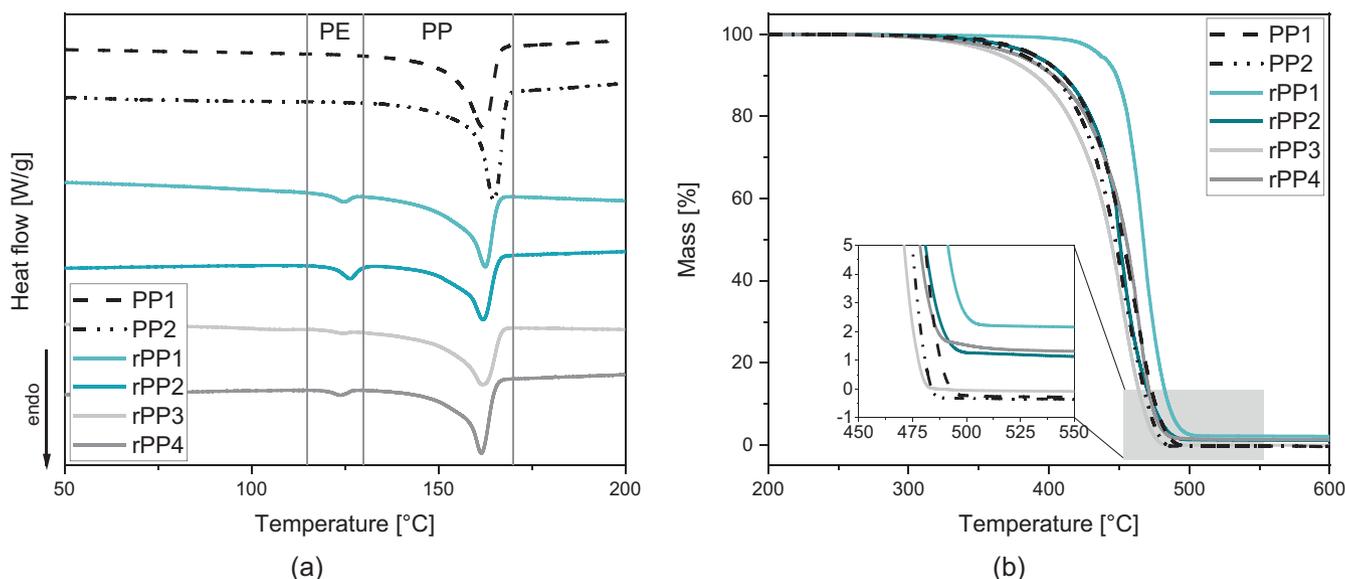
FIGURE 2 (a) Thermograms and (b) thermogravimetric analytical curves of PP1, PP2, and rPP1 to rPP4. [Color figure can be viewed at wileyonlinelibrary.com]

added from literature.[34] As expected, only one peak for PP was detected for the virgin PPs. In comparison, all PP recyclates showed two peaks, therefore a PE contamination was found. While a PE content primarily leads to inferior stiffness, several other effects are related to it. More details on the influence of PE in PP recyclates are given in further literature.[35] The lowest amount of PE was measured in sample rPP3. The other three rPPs showed a higher amount of PE contamination. The melting points (T_m) of the PE peak varied by around 3°C in a range from 123.8 to 126.5°C. The melting points of the contaminations are in the ranges of literature values of PE-LLD and PE-MD, which corroborates the assumption of PE cross-contamination. Enthalpies of fusion (ΔH_m) of the rPPs were measured in a range of 0.6 to 6.6 J/g. Melting points of the virgin PP grades and PP recyclates are located between 161.4 and 164.5°C with a slight tendency of higher melting peaks of the virgin PPs. Both PP enthalpies of fusion of the virgin PPs are around 90 J/g. It is obvious that the PP enthalpies of fusion of the rPPs

are significantly lower than those of the virgin PPs, which corresponds to a much lower crystallinity of the rPPs due to contaminations by polyethylene and also other polymeric and non-polymeric particles. The values are ranging between 63.3 and 69.1 J/g. TGA measurements revealed pyrolysis curves characteristically for PP for all samples including the rPPs, which are shown in Figure 2b. While rPP2 and rPP4 are overlapping with PP1 and PP2, rPP1 shows a better thermal stability, shown as onset temperature (T_{Onset}) in Table 3, and rPP3 a slightly worse thermal stability which is indicated with the onset of the decreasing curve. Nevertheless, for the recyclates onset temperatures between 404 and 451°C were measured which are common numbers for this material type and thus the measured thermal stability does not indicate a negative impact on possible products made from these materials.[8] No residues (m_{Rest}) in the TGA measurements were found for PP1, PP2, and rPP3. The other recyclates exhibit residual masses between 1.0% and 1.8% (see Table 3).

3.1.2 | Mechanical properties

The determined mechanical properties tensile modulus, yield stress and strain at break as well as notched and unnotched impact strengths are listed in Table 4. Especially tensile modulus and Charpy NIS are usually given in material data sheets, as they are needed for material selection. Furthermore, it is known that there is a nonlinear correlation of the material's tensile modulus and the top load of the thermoformed product.[35] Both virgin PPs PP1 and PP2 have almost similar mechanical properties where all investigated values are slightly higher for PP1 although the enthalpy of fusion and therefore the crystallinity is higher for PP2, which directly corresponds to the stiffness of the material. The absolute values are around 1900 MPa for both materials. Only a few virgin PPs with a higher stiffness are commercially available for packaging applications.[36] The tensile moduli of the four recyclates are significantly lower than those of the virgin PPs ranging from 1040 to 1240 MPa, which is in good agreement with further literature focusing on rPP.[10,27] The yield stress is also significantly lower. However, the strain at break is much higher for some of the rPPs with a maximum value of 161% for rPP3. The material rPP3 was also identified as the stiffest (high tensile modulus

and high yield stress) of the recyclates and simultaneously as the toughest (high strain at break). Although rPP1 and rPP2 should only differ in color, especially the tensile modulus of rPP2 is significantly higher. This can be explained by different input streams of the recyclates' waste stream. Nevertheless, it is obvious that a multilayer structure is necessary to achieve a certain top load. The material rPP4 shows a rather low amount of PE and therefore the mechanical properties are better than of the materials rPP1 and rPP2.

The Charpy NIS of PP1 and PP2 were measured as 3.2 and 3.7 kJ/m^2, respectively, which represent the lowest values in this comparison. All recyclates achieve higher values, which can be correlated with a certain toughness of the material due to PE and PP copolymer incorporation. The values of the recyclates range from 4.3 to 6.5 kJ/m^2, which are common values for commercial PP recyclates.[8,27] The unnotched samples revealed a reverse trend. The highest values were measured for the virgin PPs with values of 160 and 125 kJ/m^2 for PP1 and PP2, respectively. The recyclates achieved values in the range from 74 to 97 kJ/m^2. All tested samples achieved complete breaks of the specimens. While the break point is predefined for the notched samples, the unnotched samples fail at the weakest position, which is usually where a

TABLE 3 Thermoanalytical properties of PP virgin and PP recyclate materials. Values for virgin PE were taken from literature.[34]

Material	T_m [°C] PE	ΔH_m [J/g]	T_m [°C] PP	ΔH_m [J/g]	T_{Onset} [°C]	m_{Rest} [%]
PP1	—	—	162.9 ± 0.3	89.7 ± 0.2	425.2 ± 0.3	0.0 ± 0.0
PP2	—	—	164.5 ± 0.3	92.1 ± 0.1	416.0 ± 1.5	0.0 ± 0.0
rPP1	125.0 ± 0.3	3.2 ± 0.3	162.7 ± 0.2	68.4 ± 0.3	450.8 ± 0.3	1.8 ± 0.1
rPP2	126.5 ± 0.1	6.6 ± 0.0	162.2 ± 0.3	63.3 ± 0.4	423.9 ± 1.0	1.0 ± 0.0
rPP3	124.4 ± 0.1	0.6 ± 0.1	161.8 ± 0.1	69.5 ± 0.8	404.2 ± 0.3	0.0 ± 0.0
rPP4	123.8 ± 0.1	2.1 ± 0.0	161.4 ± 0.1	69.1 ± 0.3	417.8 ± 0.8	1.2 ± 0.1
PE-LD	105–115	—	—	—	—	—
PE-LLD	120–130	—	—	—	—	—
PE-MD	120–130	—	—	—	—	—
PE-HD	128–136	—	—	—	—	—

TABLE 4 Mechanical properties determined from tensile test and Charpy impact test of all investigated materials.

Material	Tensile modulus [MPa]	Yield stress [MPa]	Strain at break [%]	Charpy NIS [kJ/m^2]	Charpy UIS [kJ/m^2]
PP1	1910 ± 20	37.9 ± 0.1	48 ± 12	3.2 ± 0.1	160 ± 15
PP2	1880 ± 10	37.3 ± 0.1	18 ± 3	3.7 ± 0.1	125 ± 7
rPP1	1040 ± 10	25.8 ± 0.1	56 ± 17	6.5 ± 0.2	85 ± 6
rPP2	1180 ± 10	26.4 ± 0.1	67 ± 22	5.9 ± 0.3	97 ± 19
rPP3	1240 ± 10	30.1 ± 0.1	161 ± 17	4.3 ± 0.5	94 ± 24
rPP4	1190 ± 10	27.1 ± 0.1	86 ± 18	5.5 ± 0.3	74 ± 24

macroscopic contamination is incorporated in the material. A previous study has shown the effect of various macroscopic contaminants on the mechanical failure behavior of PP.[10] Therefore, materials with no defects like PP1 and PP2 deliver higher values than recyclates with most probably containing defects. A graphical illustration of the property profiles including data from tensile test and Charpy test is shown in Figure 3.

3.2 | Testing of modification strategies at product level

Multilayer cups were produced using all 4 PP recyclates as a core layer and PP2 as top layers. Due to the fact that the stiffness is of the recyclates are significantly lower than those of the standard materials and that there is a correlation between tensile modulus and top load, the production of monolayer recyclate cups was not carried out. In Table 5, the results of product tests are shown for the control cup and the multilayer cups containing rPP with the setups c1, c2, c3, and c4. The geometrical target value for the shrinkage diameter is set to 100%. Moreover, the top load of the control sample made from the
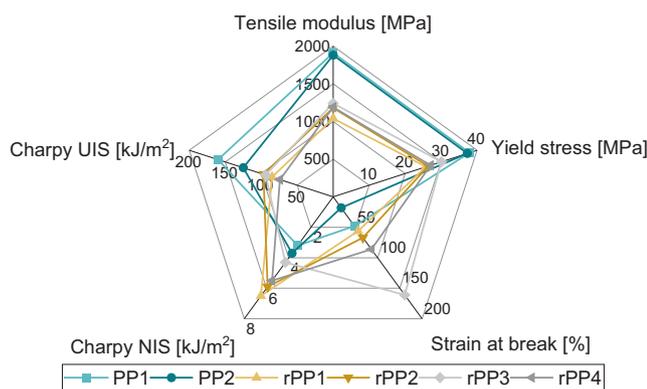
FIGURE 3 Material property profiles of PP1, PP2, and rPP1 to rPP4. [Color figure can be viewed at wileyonlinelibrary.com]

TABLE 5 Shrinkage diameter and top load as a relative value based on a comparison with the target values for cups from films of the reference material and from multilayer films with all four recyclates.

Cup	Shrinkage diameter [%]	Top load [%]
control	99.73	100
c1	100.23	75.8
c2	100.30	88.2
c3	100.24	111.4
c4	100.31	95.3

virgin PP is set to 100%. The shrinkage diameter of the control cup is the lowest of all investigated cups as shrinking is not restricted by PE inclusions and different PP domains. Cups c1, c2, c3, and c4 achieved values above the target value, with the largest deviation for cup c4 and the smallest for cup c1. Although the deviations appear to be very small, a high degree of reproducibility is required to ensure an optimum fit of the lid for closing the thermoformed cup. The possible deviation margin is higher for lower values than for higher values.

When comparing the top loads, cups c1, c2, and c4 achieved significantly lower values by 5% to 25% than the control cup, but cup c3 even exceeded the reference value by more than 10%. No target ranges were set for this product, as the ranges are dependent on the envisaged product. Independent of the product an assumed tolerance limit of ±10% would be always valid and in this case, the cups c1, c2, and c3 would fail this test. Although rPP3 has a lower stiffness than PP1, the comparable performance in top load is possible due to the reinforcing top layers with virgin PP. It seems that not only the stiffness of the individual material is influencing the product stability but also the multilayer film setup itself. Another reason could be the unknown amount of in-house recyclate material in the control cup made from edge trims and punching grids, which could have a negative influence on the performance of the control cup. Nevertheless, it seems like the right amount of top layer material was chosen to get close to the reference value. Regarding top load results, the same ranking is achieved as for the tensile moduli of the pure materials. As the top layers are kept constant in this comparison, a non-linear dependence of stiffness obtained from tensile test at material level and top load test at product level exists. A similar comparison of stiffness and product performance for an injection molded multilayer box was found by Gall et al.[10] During thermoforming a high melt strength is necessary to obtain satisfying results. An indirect relationship of melt strength and the MFR of polypropylene are known.[37]

In Figure 4, the influence of the thickness of the top layer and the influence of added virgin PP to the rPP in the core layer is shown according to strategies (i) and (ii), respectively. From left to right the absolute recyclate content decreases. The left two bars of both diagrams represent the films with thinner top layers c1_s and c1_sb, which corresponds to a higher recyclate content in the cup. The right two bars are the samples with a thicker top layer (c1) and additionally a blend with virgin PP in the core layer (c1_b). The recyclate content of the cup was slightly decreased for both setups following strategy (ii) by mixing 15% PP1 to the recyclate in the core layer to adjust the melt flow rate of both layers according to
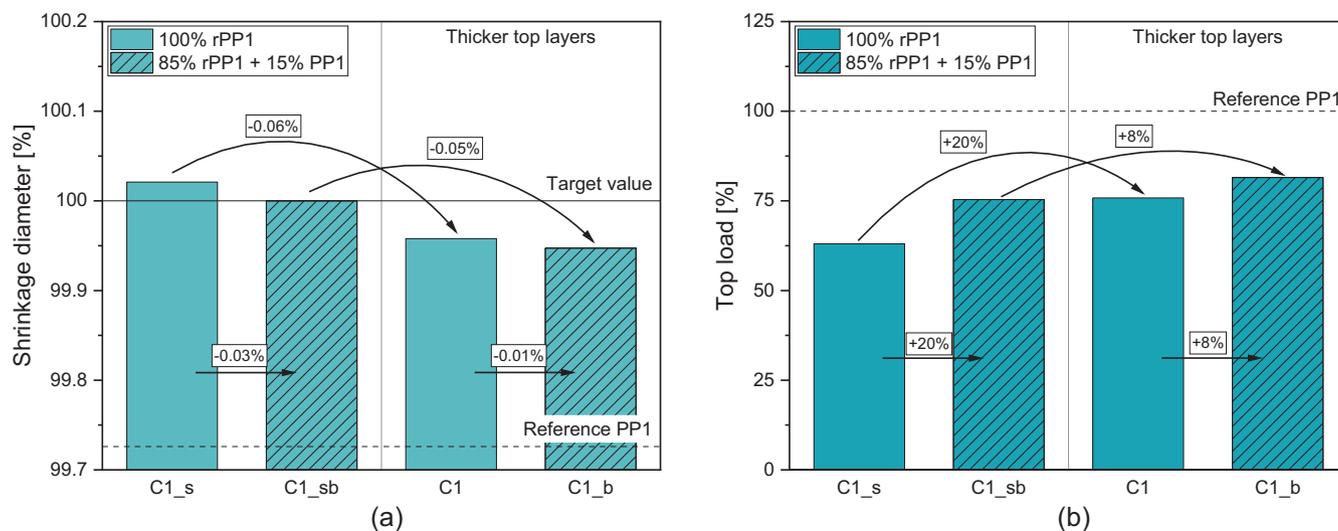
FIGURE 4 Effect of layer structure and added material on (a) shrinkage diameter and (b) top load (decreasing recyclate content from left to right). [Color figure can be viewed at wileyonlinelibrary.com]
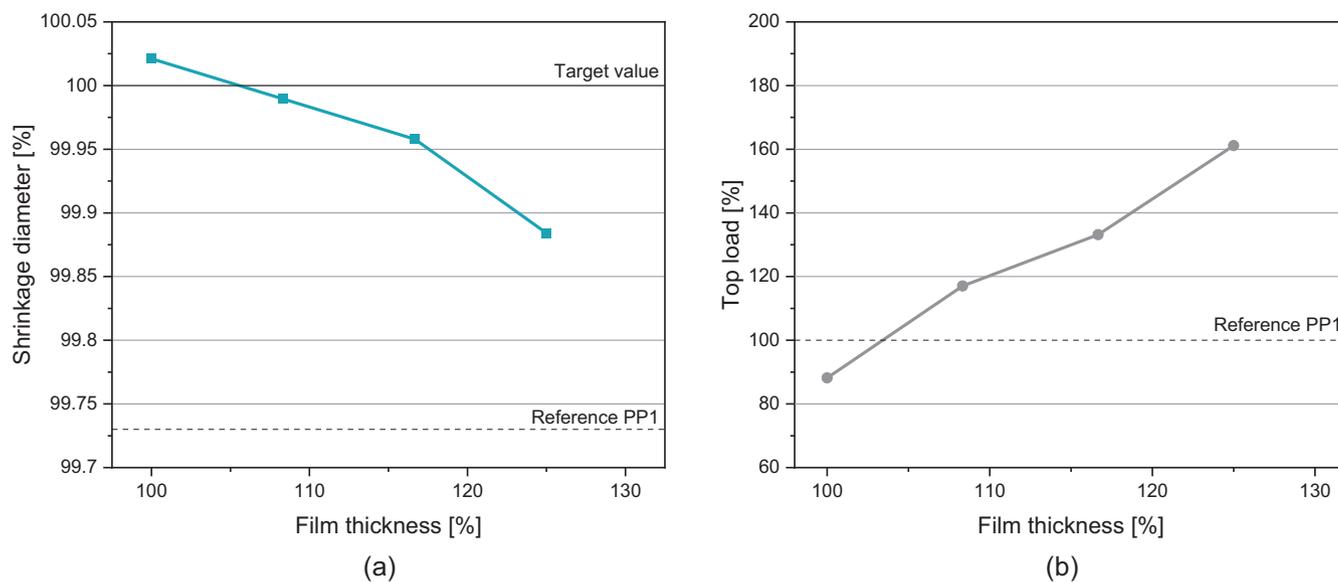
FIGURE 5 Influence of absolute film thickness of PP2-rPP2-PP2 on (a) shrinkage diameter and (b) top load. [Color figure can be viewed at wileyonlinelibrary.com]

Traxler et al. (2022) which is represented by the patterned bars.[28] The curved arrows on top of the bars indicate the change of the shrinkage diameter (Figure 4a) by increasing the thickness of the top layers while the material in the core is kept constant. In both cases, the change in shrinkage diameter is approximately the same with values of 0.06% and 0.05%, respectively. It does not matter whether the core layer has been diluted with virgin PP or not. Diluting the recyclate in the core layer with virgin PP (indicated by the straight arrows in Figure 4a) results in a slight decrease of the shrinkage diameter, although this is more pronounced for the thinner top layer, as

there is more material in the core layer overall. Percentage changes of 0.03% and 0.01% were calculated.

Figure 4b shows the influence of the different material structures on the top load. It can be summarized that the top load increases with a lower recyclate content. However, the value of the control cup is significantly higher than those of all multilayer cups made with rPP1. By adding virgin PP to the rPP in the core layer or increasing the cover layer thickness, an improvement in the top load of approx. 20% was achieved (comparison of c1_s with c1_sb and c1). Accordingly, the same value was measured for cup c1_sb and cup c1. When comparing

cup c1 and c1_b, an improvement of 8% was achieved by adding virgin PP to the core layer, whereas the same structural adjustment of the film with thinner top layers resulted in an improvement of around 20%, as the absolute amount of virgin PP in the overall multilayer structure is higher compared to the other film.

For the evaluation of strategy (iii) the change of the shrinkage diameter over the overall film thickness increase is plotted in Figure 5a. The overall thickness of the multilayer film was increased by up to 25%. The shrinkage diameter decreases with increasing film thickness. The temperature of the cup after demolding remains high for a longer time than for thinner cups. Therefore, crystallization processes take longer which leads to a higher crystallinity and a higher shrinkage of the material.[38] It is known that a higher crystallinity leads to lower shrinkage diameters.[35] This demonstrated behavior is in good agreement with the results presented in this work and gives a clear indication of the relationships of morphology and product properties. Nevertheless, all values are within the tolerance range. At 100% thickness the shrinkage diameter is above the target value. For the three other tested film thicknesses, the value drops below the reference value of the control cup. All values are located above the value of the control cup made from the virgin PP. Up to 117% of the reference film thickness; the decrease in shrinkage diameter is linear, while for 125% a decrease is even more pronounced.

At the product level, components become more stable due to a higher material input in the form of thicker walls, which results in an increased moment of inertia. The failure of the cup under compressive load corresponds to buckling. Figure 5b shows the results of the top load tests where the value of the PP1 control cup is set to 100%. The intersection of the gray curve with the dashed horizontal line indicates the additional wall thickness required to achieve the same stability as the control cup. In this case, this is an additional wall thickness of 3%. In total, the top load can be increased by 83% by using 25% more material. However, when evaluating this multilayer structure economically, it may not be competitive with other solutions.

4 | SUMMARY AND CONCLUSION

Four different commercially available polypropylene recyclates (rPPs) were selected for thermoforming tests and processed into core layers of multilayer cups, the top layers of which consisted of virgin polypropylene (PP). Three strategies were defined and tested to enable constant and adequate products properties. Hence, various parameters of the cup structure were varied, such as (i) the share of top layers, (ii) the material composition in the core layer, and (iii) the overall thickness of the extruded film. Thermoanalytical characterizations and mechanical tests were carried out as basic characterization for a quality assessment at the material level. The thermograms showed pronounced polyethylene peaks and significantly lower PP enthalpies of fusion for all rPPs compared to the virgin PPs. Furthermore, pyrolysis residues of up to 1.8% were detected in the rPPs. The mechanical properties of the rPPs in terms of stiffness were significantly below those of the reference virgin PPs, justifying the use of multilayer structures. Impact strength testing of the rPPs resulted in higher values for notched specimens and lower values for unnotched specimens compared to virgin PP. By implementing the three predefined strategies at the product level to adjust the product properties of multilayered cups, the property profiles of the multilayer cups led to values closer to the target values of thermoformed cups made from a virgin PP only. However, strategy (iii), the increase in overall thickness, leads to an uneconomical additional material consumption, which should be avoided. In general, with the selected materials and the considered strategies for multilayer thermoformed products, it was possible to produce multilayer PP cups with a high recyclate content and with adequate product properties. These strategies for product adjustments could lead to higher recyclate contents in the packaging industry as the recyclate is sealed from the packed products and the environment, which simplifies the use of recyclate in various products. Since the proposed solution of using multilayer structures does not use any material mixtures, but only combines virgin and recycled material of the same material type, there is no loss in terms of further recyclability and sustainability.

CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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