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Sustainable thermoplastic composite parts through back injection molding of UD-tapes with recycled PET materials

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Abstract

The demand for advanced and sustainable materials is increasing as industries aim to reduce their carbon footprint. Polyethylene terephthalate (PET) bottle recycling is a prime example of a successful circular economy in the field of plastic products. However, the recycling process often results in materials that no longer meet the required specifications and cannot be returned to the cycle.

In the field of injection molding, there is an opportunity to use recycling PET (rPET) for technical parts currently made with other materials. Combining rPET with continuous fiber-reinforced tapes in the back injection molding process shows promise for producing rigid composite parts sustainably. Initial investigations indicate comparable mechanical properties to today favored technical materials like polyamide (PA).

Introduction

The growing emphasis on sustainability, driven by new laws, regulations, and consumer demand, is pushing plastics processing companies to reduce the carbon footprint of their products without compromising on performance or cost. Incorporating recycled materials presents a significant opportunity to achieve this goal, but availability and quantity constraints hinder its widespread adoption. A material that is fulfilling this requirement and is available in large quantities is recycled polyethylene terephthalate (rPET). According to data from 2013, rPET accounted for 14.4% of the total plastic waste in the United States. This figure places rPET right after the three most prevalent polymers in the packaging waste stream, namely polypropylene (PP), low-density polyethylene (LDPE), and high-density polyethylene (HDPE) [1].

PET is a common thermoplastic polymer with outstanding mechanical properties that is used in various applications. However, there are some limitations using PET for injection molding due to its tendency to crystallize, sensitivity to moisture, and specialized equipment and processing techniques required. This can increase production costs and requires a lot of processing knowledge.

Today there are different sources for rPET. PET bottle recycling in Switzerland is a prime example of a successful circular economy in the field of plastic products, demonstrating the potential to reduce CO_2 emissions. In 2021 more than 82% of the 42’393 tons of PET material was recycled [2]. However, the recycling process often results in materials that no longer meet the required specifications and therefore cannot be returned to the cycle (e.g. color, contamination).

Today these materials are either spun into polyester fibers or used for energy recovery and remain practically unused for technical parts. The processing is often even more difficult than with virgin PET. There are only few examples using rPET for technical parts, like the investigation of the suitability of rPET material for a canopy strip in a commercial vehicle [3].

Beside the residuals from the PET bottle stream there is another source for rPET, which is ocean-bound plastic material collected from the sea or beach, which will then be recycled. This upcycling process produces up to 80% less CO_2 emissions than producing virgin plastics, according to #tide Ocean [4].

In the field of injection molding, there is a significant opportunity for these materials, especially with the increasing trend towards using or switching to post consumer recycling (PCR) materials to reduce the carbon footprint. By adding suitable additives (nucleating agent, minerals, short fibers) to rPET, it can also be used to produce technical parts, currently made with polybutylene terephthalate (PBT), polyoxymethylene (POM) or polyamide (PA) with comparable cycle times in injection molding [5].

Combining these rPET compounds with unidirectional fiber composites tapes (UD-tapes) in the back injection molding process presents a promising opportunity for not only producing technical parts, but also producing highly rigid sandwich composite parts sustainably. Back injection molding of UD-tapes offers numerous benefits, such as improved mechanical properties, higher productivity, and lower material wastage. By using unidirectional fibers, the composite can be engineered to be highly anisotropic, with exceptional strength and stiffness along a single axis.

The resulting composites have shown significant improvements in terms of their mechanical properties, such as tensile and flexural strength, impact resistance, and fatigue life [6]. The combination of unidirectional fiber composites and injection molding has also enabled the production of complex geometries and parts with high precision and repeatability. These benefits have led to the adoption of this technology in various applications, such as automotive parts, sporting goods, and industrial equipment and replace conventional composite applications made of thermoset materials. A practical example for this can be fins for surf boards for professional use. Previously, these surf fins were hand-produced in China using a classic resin transfer molding process. However, it was possible to replace this process with injection back molding of UD-tapes, allowing for production to be transferred to Switzerland [7].

Currently, there are several types of UD-tapes with thermoplastic matrix available on the market. Some of the most used matrix materials include PA and PP, wherein the reinforcing fiber is carbon, glass or aramid. But PET and rPET are also increasingly entering the market as matrix materials, which enables the possibility to use rPET for partially highly reinforced parts as well.

The fibrous material and matrix of the tape, as well as the material for injection molding, are available from different suppliers in various combinations. Choosing the right combination for the current application is not trivial, as many factors have to be considered. The ability to combine different materials and adjust parameters such as fiber orientation, tape thickness, and reinforcement of the injection molding material allows for adjustable product properties. Thus, with the same process in the same mold, different stiffnesses or strengths can be achieved. Alternatively, by changing the fiber angle on the tape, a specific deformation behavior can be favored or prevented, which is very attractive for stressed components, but also makes their design difficult [8].

Material selection

There are different suppliers which offer UD-tapes with a PET matrix. Often there is a choice between PET-A (amorphous) and PET-G. PET-G is a glycol-modified adaptation of PET which has also amorphous structure and offers some advantages, such as printability. This could be interesting for some applications. Currently, a lot of research work is being done on the development of tapes with rPET matrix. Often, these tapes are not commercially available yet.

Most of these PET-tapes contain glass fibers as reinforcement, just a few could be found with carbon (CF) as fiber material. The tapes that were considered for experimentation are shown in Table 1.

Table 1. UD-tape materials

Material	Type	Fiber content in %	Supplier
PET-G / CF	CF-PETG-43-100-190	43	A+ Composites GmbH
PET-A / GF	Polystrand IE 5840B	58	Avient Corporation
rPET / GF	Polystrand IE R-5840B	58	Avient Corporation
PA12 / CF	CF-PA12-41-102-202	41	A+ Composites GmbH

The material for back injection molding is typically chosen based on its compatibility with the UD-tape and the specific performance requirements of the final product. Reinforced materials are commonly used in combination with tapes for two reasons. On one hand to enhance the strength, stiffness, and durability of the produced part. On the other hand, because of its lower shrinkage compared to unreinforced materials. The tape prevents shrinkage at the location of reinforcement and the goal for the finished part is to be free of internal stresses and not to warp. Therefore, a reinforced material is more suitable.

The injection molding materials for experimentation are shown in Table 2.

Table 2. Back injection molding materials

Material	Type	Fiber content in %	Source	Supplier
rPET GF20	G2040PET-00G20PH	20	Ocean bound	#tide Ocean Material
rPET GF50	-	50	Ocean bound	IWK
PA12 GF20	Grilamid LV-2H	20	Virgin	EMS-Chemie AG
PA12 GF65	Grilamid LV-65H	65	Virgin	EMS-Chemie AG

The rPET GF50 material was compounded in the laboratory of the Institute for Materiel Technology and Plastics Processing (IWK) which belongs to the Eastern Switzerland University of Applied Sciences. This material is not commercially available by now.

Experimentation

The experimentation was done with two different part geometries. As a first step, basic investigations were carried out on a simple test geometry. These results were then transferred to practical geometry.

The specimen for the basic investigations was a simple plate with UD-tapes on both sides (sandwich structure), as displayed in Figure 1. The dimensions of the plate are 60 x 80 x 4.3 mm³.

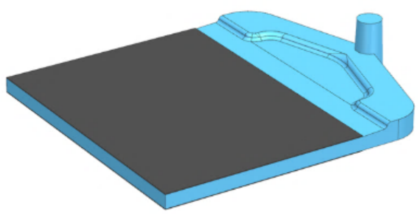

Figure 1. Sandwich plate for basic investigations

In a first step the effect of preheating the tape with an infrared heater was evaluated. But even without preheating it turned out that the adhesion with the PET material combinations is much better than expected, respectively as known from the PA materials. Very good adhesion between the tape and the back-molded material was achieved even with only partial filling of the cavity.

Moreover, the preheating process caused distortions on the tape, which remained visible even after back injection molding process and it became challenging to keep the tape in place within the cavity. For this reason, it was decided to carry out the tests without preheating the tapes.

To evaluate the optimal process parameters for each material / tape combination a trial plan based on D-optimal experimental design was developed, wherein the parameters *melt temperature, injection speed* and *holding pressure* varied. The parameters of each variant are displayed in Table 3.

Table 3. Variants of the D-optimal experimental design

Variant	Melt temperature in °C	Injection speed in cm^3/s	Holding pressure in bar
V1	290	45	350
V2	280	20	250
V3	280	65	450
V4	300	20	450
V5	300	65	250
V6 (=V1)	290	45	350

For comparison, samples of a PA GF20 reference variant with the parameters 300 °C melt temperature, $60 cm^3/s$ injection speed and 350 bar holding pressure were produced.

Following that, injection molding trials using the practical geometry were conducted. The surf fin, as illustrated in Figure 2 and previously described, served as part for these trials.

The back injection molding material was changed to incorporate grades with a higher level of reinforcement for this phase of experimentation.

Figure 2. Surf fin used as practical geometry (rPET)

Results

The mechanical characterization of the plates was done in 3-point bending test according to DIN EN ISO 14125:2011-05. Figure 3 (at the end of this paper) shows the force at a deflection of 1 mm for the different variants of the D-optimal experimental design. These basic investigations were performed with the back injection molding materials with a reinforcement level of 20%.

The higher stiffness of the variants with CF tapes is obvious. Additionally, these variants just show a slight influence of the setting parameters. Higher melt temperature and faster injection tend to result in a higher flexural modulus. However, the differences are very small and confirm the findings from first adhesion tests, that adhesion is similar in all variants.

For the variants with GF, there are practically no differences observed within the variants. Furthermore, when rPET is used as the matrix material for the tapes, there is no significant difference compared to tapes with PET virgin material. The scattering tends to be even lower with rPET tapes.

Interestingly, the stiffness of the PET-G/CF tape variants is up to 9% higher than the one of the polyamide reference variant. According to model calculations for the stiffness of a simple sandwich structure, a more or less similar stiffness was expected. However, this could be attributed to the better adhesion between the back injection molded material and the tape with PET.

Figure 4 shows the force / deflection curves for different PET-tapes compared to the PA material combination as reference. The stiffer behavior of the PET-G/CF variant is obvious in this diagram. It can be observed that a first damage occurs at a deflection of approximately 1.5 mm (kink in the curve). This could be either delamination or fiber fracture.

In comparison, the polyamide variant shows damage already at a lower deflection (around 1.3-1.4 mm). This further favors the use of the sustainable material combination.

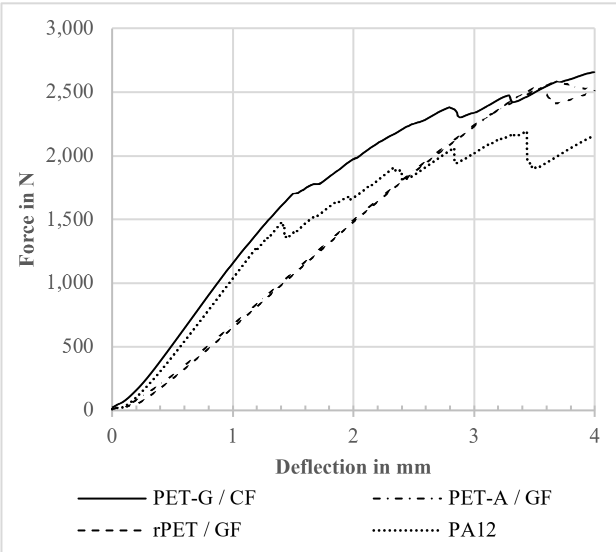

Figure 4. Force / deflection curves in a 3-point bending test

To summarize the basic investigations, it can be said that the sustainable material combinations offer mechanical properties that are on par with, or even slightly superior to, currently favored materials such as PA12.

The surf fins are tested on a tensile testing machine to determine their stiffness. Therefore, the surf fin is fixed on one side and a force of 75 N is applied on four different positions according to figure 6. The deflection is measured at each of these points (Figure 5).

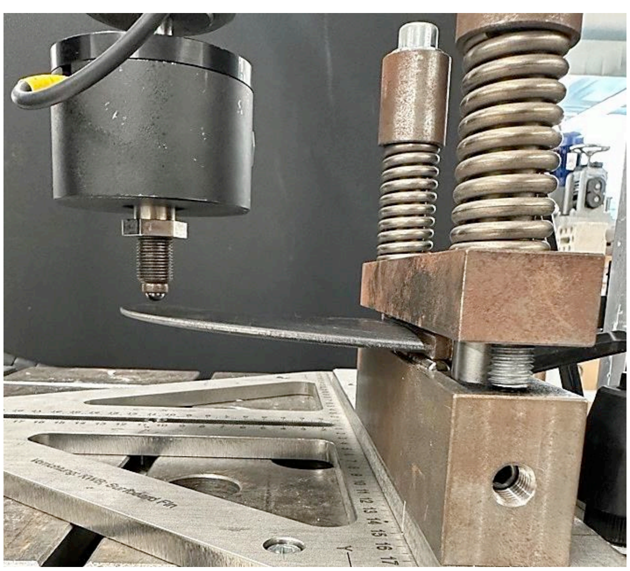

Figure 5. Set-up for local stiffness measurement

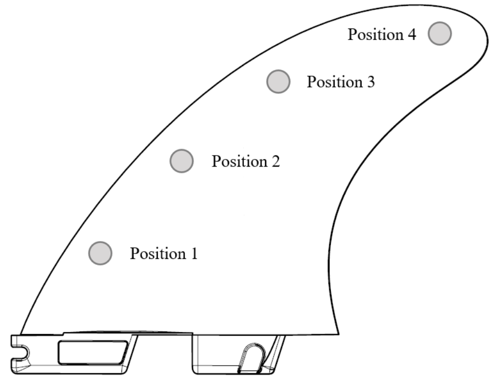

Figure 6. Measuring positions for determining the local stiffness of the surf fin

Table 5 presents the local stiffness values for the rPET surf fin in comparison to a PA12 fin. Despite the PA12 variant incorporating 65% glass fibers in the back-injected material, the rPET material, which contains 50% glass fibers, demonstrates an almost equal performance.

Table 5. Local stiffness comparison, surf fin

Position	rPET GF50	PA12 GF65	Ratio
1	233.4 N/mm	210.7 N/mm	111 %
2	75.7 N/mm	80.3 N/mm	94 %
3	35.4 N/mm	35.9 N/mm	99 %
4	10.6 N/mm	10.7 N/mm	99 %

Depending on the measuring position, an increase in stiffness of up to 11% could be achieved. In measurement positions 2 to 4, the local stiffness of the rPET variant falls slightly short, though the decrease is minimal.

Utilizing rPET leads to a higher weight for the parts compared to PA12. Nonetheless, the slightly improved mechanical properties allow for the use of a material with lower glass fiber (GF) content in the back injection molding process. In the case of the surf fin example, we note only a marginal increase in weight of 4%. Considering its exceptional adhesion, foam injection molding emerges as a potential solution to even reduce the weight compared to PA12.

Conclusions

The results highlight the significant potential of utilizing sustainable thermoplastic composite parts, achieved through the back injection molding of UD-Tapes with recycled PET materials. This approach not only proves to be feasible but also stands out as an environmentally friendly option. The produced parts have potential for applications across various industries such as automotive, energy, sports, and leisure.

Which exact applications such material combinations can be used for depends not only on the achievable mechanical properties but much more on the given environmental conditions.

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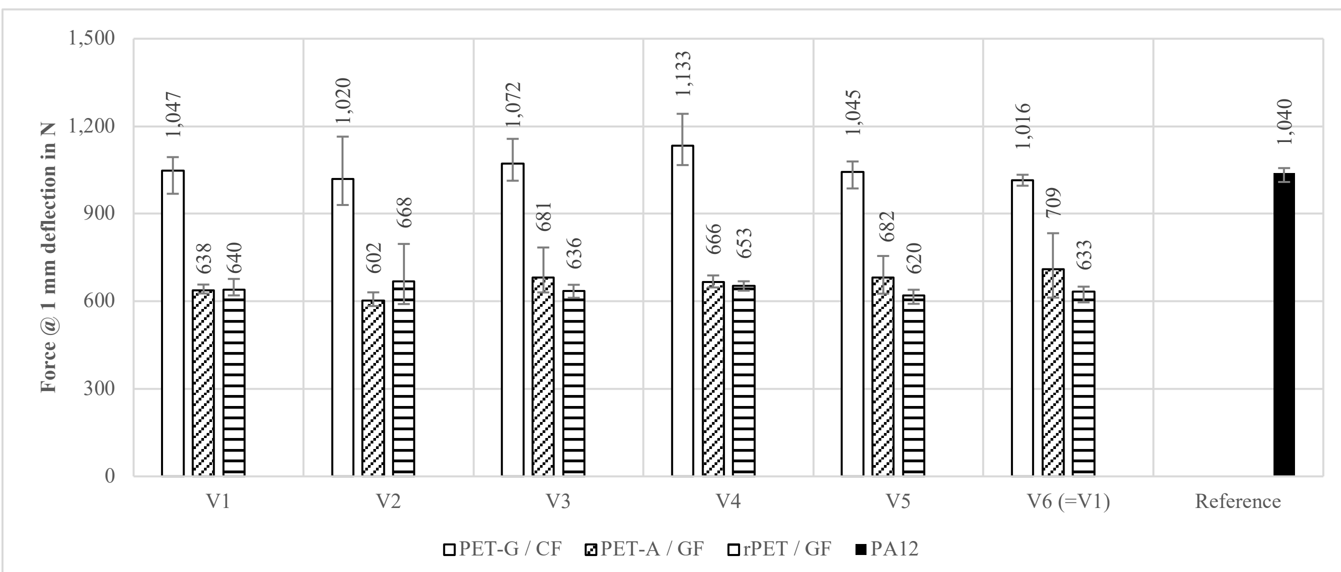

Figure 3. Force at 1 mm deflection for the different variants of the D-optimal experimental design, compared to PA12