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Mechanical Recycling of Waste from PET Bottles and Glass Polyester Composites

Joanna BRZESKA¹, Mariola JASTRZĘBSKA¹, Magda MORAWSKA¹, Maciej SIENKIEWICZ²

- ¹ Department of Industrial Products Quality and Chemistry, Gdynia Maritime University, Poland
- ² Department of Polymer Technology, Gdansk University of Technology, Poland

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Abstract

Composites are used in many areas of the economy, especially in transport and green energy production. As a consequence, a large amount of composite waste is generated, which is difficult to recycle. In this study, an attempt was made to use the simplest and cheapest mechanical method to process post-consumer and post-production waste of coloured polyester and glass polyester composite, respectively. Composites were prepared by injection moulding, adding ground unsaturated polyester with glass fibre to shredded poly(ethylene terephthalate) (rPET). The effect of adding the glass polyester recyclate on selected mechanical (tensile and impact strength), thermal (using differential scanning calorimetry), surface (contact angle), and structural properties (using the Fourier infrared spectroscopy and optical microscopy) was determined. The research has demonstrated that it is possible to obtain in a simple way high quality, aesthetically pleasing and weather resistant landscaping products, such as fences and road posts, litter bins, benches, etc., from post-consumer coloured PET bottles and post-production waste glass polyester composites. Nevertheless, in order to obtain composites with better mechanical properties, the recyclates used need to undergo additional processing, e.g. by adding compatibilisers.

Keywords

composite waste, post-consumer PET, mechanical recycling, thermoplastic composite.

Introduction

To improve the quality of the natural environment –and, consequently, the standard of living – it is essential for individuals to take responsibility for proper management of plastic waste. In some cases it is easier (such as for linear polyesters), whereas for some other plastic materials it is almost impossible to process and reuse them (e.g. cross-linked polymer resins, composites, etc.). Thus, there are many options for recycling the used polyester, i.e. poly(ethylene terephthalate) (PET) bottles, but the disposal of glass fibre reinforced resin composites, such as those used in yacht construction, is severely limited (Fig. 1). In this study the authors have decided to use glass polyester recyclate to reinforce the matrix from poly(ethylene

Corresponding author: Joanna Brzeska – Department of Industrial Products Quality and Chemistry, Gdynia Maritime University Morska, 81-225, Gdynia, Poland, e-mail: j.brzeska@wznj.umg.edu.pl

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terephthalate) waste. The main objective of this work is to find a solution to the research problem of simple management of polyester glass composite waste. For this purpose, mechanically recycled green-dyed PET bottles (rPET), as well as recyclates from ground-up waste post-production yacht hulls, were used to obtain thermoplastic composites for small architectural objects. The preliminary tests were to show whether the resulting product would meet the visual requirements and, above all, whether it would be possible to obtain a product with appropriate mechanical properties in a simple way without additional modifications (e.g. introducing a compatibiliser). The research hypothesis assumes that a material suitable for small architectural objects can be obtained in a simple way. It is known that the main application area for thermoplastic composites is the transportation sector, which represents almost two thirds of the market (65.6%). Together with electrical and electronic applications, this resulted in a market share of almost 90% by 2023 (Witten & Mathes, 2024). Small architectural objects could also constitute a good market for products from rPET composites with glass polyester recyclate.

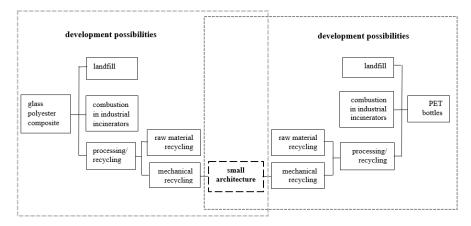


Fig. 1. Scheme of waste management options for glass polyester composites and poly(ethylene terephthalate) bottles

Literature review

The volume of the global polymer composites market (thermosets and thermoplastics) in 2023 was 13×10^6 Mg (Witten & Mathes, 2024). The share of Europe in the global market decreased by 8% compared to 2022 and reached around 20% of the global market. Germany remains the largest market among the regions reviewed with a share of more than 19.4% of the total market. Eastern Europe ranks second with a market share of 19% and a volume of 0.204×10^6 Mg annually. This region includes the following countries: Poland, the Czech Republic, Hungary, Romania, Serbia, Croatia, Macedonia, Latvia, Lithuania, Slovakia, and Slovenia. Glass fibre reinforced polymer composite systems still account for more than 95% of the overall market. The total production volume of glass thermoset composites (mainly polyester and epoxy resin) amounted to 1.073×10^6 Mg in 2023 (Witten & Mathes, 2024). Therefore, this material group represented 43% of the total market in Europe. The construction sector (36.3%) remains the most important area of thermoset composite application, followed by the transportation sector (28.9 %) and applications in the electrical and electronic sector (17.2%) (Witten & Mathes, 2024). Polyester resins are widely used in many applications because of their low price and good chemical properties. The Polish yacht and recreational boating sector is an important branch of the economy in Poland - around 25 thousand boats were produced from composite materials in 2022 (Polish Investment & Trade Agency, 2023). The annual amount of post-production scrapped glass polyester reaches about 2×10^3 Mg. Moreover, their reinforcement, e.g. with glass fibre, results in obtaining a material with exceptional mechanical properties. Therefore, such composites are used in applications

that require high mechanical strength combined with low weight, as in the construction of yachts, wind turbine blades, and similar structures. There are also 20×10^3 Mg of glass polyester composite objects at the end of the yacht's working life. These excellent properties of polyester composites with glass fibre are largely due to the high degree of cross-linking of the polyester matrix and strong interaction with a glass filler. However, the same characteristics make them extremely difficult to recycle. This issue has been examined extensively for many years, and several methods of recycling glass reinforced composite waste have been described (Krauklis et al., 2021; Gonçalves et al., 2022). Fundamentally, two recycling technologies were investigated and two potential waste management solutions were found: mechanical recycling and raw material recycling, which includes chemical recycling and coprocessing. Nevertheless, raw material recycling facilities are extremely rare due to their complexity and cost.

Most glass reinforced unsaturated polyester composites are produced for a specific purpose, and there is no possibility of reusing them in other applications. On the other hand, waste (off-cuts parts) that is generated during production can be reused. In mechanical recycling, the composite waste is ground into finer fractions, which can then be used as a filler or reinforcement in virgin or recycled composite resin (Meira Castro et al., 2013; Palmer et al., 2009).

Another polymer waste generated in large quantities is poly(ethylene terephthalate). Its popularity in such applications as: beverage and water bottles, fibres, and insulating materials is due to its good tensile and impact strength, transparency, thermal stability, and chemical resistance. In 2022, the global size of the poly(ethylene terephthalate) market was 25.47×10^6 Mg, of which 2.94×10^6 Mg was produced in Europe (Statista Research Department, 2024). This

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represented 5% of the 58.8×10^6 Mg of the total European plastic production. 3.7×10^6 Mg of PET waste was converted into new plastic products (Plastics Europe, 2024). A vast majority of PET packaging collected were bottles, accounting for 2.8×10^6 Mg (Independent Commodity Intelligence Services, 2022). In 2022, the recycling collection rate for beverage bottles reached 75%. In the same year in European countries PET waste was reprocessed into flakes by 140 companies. Currently, recycled PET is used mainly in fibre and bottle production. A significant number of food-grade and non-food-grade bottles are made from recycled PET. Clothing fibres are also made largely of recycled materials. Nine clear plastic water bottles can be used to make one T-shirt, while 25 plastic bottles can be used to make a pair of leggings. Some other uses of recycled PET include apparel for outdoor sporting wear, such as thermal undergarments, knapsacks, sweaters, and socks (MarketsandMarkets[™]. 2024). Mechanical recycling is the most common and cost-effective method. These recycling processes reduce the need for virgin PET, thus reducing greenhouse gas emissions, energy consumption, and the depletion of natural renewable resources. This involves shredding and melting the PET material to produce recycled PET (rPET) pellets. Although mechanical recycling of PET waste is a simple process and probably the most ecologically acceptable (apart from reusing), there is a group of waste materials that cannot be subjected to such a process. These are coloured or multilayered trays, foils, bottles, etc. It is estimated that 20% of the bottles contain various coloured PET. The higher the amount of colouring (and darker colour), the fewer the applications for which the rPET can be used. Therefore, as the colouring increases, more material is lost to successively darker coloured products. In their case, chemical depolymerisation is more important, leading to the creation of initial monomers, devoid of dyes and other additives. The recycling of polyester packaging, such as coloured and multilayer trays and films, remains a significant challenge for mechanical processes. Therefore, chemical depolymerisation is popular. Consequently, colourless recycled PET flakes have higher prices in Eastern Europe (ICIS 2024).

Recently, the market for thermoplastics composites, including PET, has expanded reaching a total volume of 1.423×10^6 Mg in Europe in 2023 (Witten & Mathes, 2024). It is observed that after adding short glass fibres to polymers, they significantly improve their properties compared to those of nonreinforced materials. In particular, there is a positive influence on the modulus of elasticity and stiffness of the materials. The glass fibres contained are generally less than 2 mm in length. Short glass fibre reinforced thermoplastics remain by

far the largest segment in the composites industry $(1.3 \times 10^6 \text{ Mg})$ (Witten & Mathes, 2024). Research on thermoplastic composite materials manufactured from recycled PET is being continuously conducted (Giraldi et al., 2005; Muringayil et al., 2024). For example, Sim et al. (2021) demonstrated that mechanical characteristics of recycled PET glass fibre thermoplastic composites were improved by increasing the fibre content.

Materials & Methods

Tested Materials

In this study, glass polyester recyclate was used as a filler in the PET matrix achieved from the regrinding of bottle waste flakes.

Glass polyester recyclate

Offcut samples were collected from the production of hulls for yachts. Glass fibre-reinforced, cold-cured unsaturated polyester composite waste was ground in a shredder manufactured in Kubala Sp. z o.o. (Ustroń, Poland). The recyclate was a mixture of cured polyester resin particles and glass fibre. Recyclate grains were smaller than 0.75 mm.

Green PET-flakes

The recycled PET in the form of flake (PET flakes) was obtained from washed and shredded waste originating from green soft drink bottles. The PET-flakes were irregular in shape and were dried in an oven at 130°C for 24 h. The use of such a recyclate tends to cause slight discolouration and minimal reduction in the clarity of PET.

$Composites \ of \ rPET \ with \ glass \ polyester \\ recyclate$

To obtain ready-made shapes for strength and impact tests, PET-flakes were injected together with glass polyester recyclate on a Battenfeld HM 45/130 injection moulding machine. The volume of one injection cycle was $32~{\rm cm}^3$. The process was carried out with the following temperature profile: $280-270-260-250^{\circ}$ C. Ten ready-made shapes were obtained from each series for further tests. The injection moulding machine parameters included an injection pressure of 500 bar applied for 5 seconds.

In this study, glass polyester recyclate in different amounts (2.5; 5; 7.5; 10 wt.%) was added before injection into the rPET matrix in composites, giving rPET 2.5%, rPET 5%, rPET 7.5% and rPET 10%. A sample

without recyclate (rPET 0%) was also prepared for comparison. The tensile strength samples were prepared according to EN ISO 527: 2020 with dimensions $4 \times 10 \times 150$ mm and for an impact test according to EN ISO 179-1:2023 unnotched 4×10 mm. Samples for DSC and microscope were cut from injected materials.

A scheme of the individual stages of obtaining rPET modified with glass polyester recyclate is shown in Figure 2.

Methods

Optical microscopy

Optical microscopy (OM) micrographs were taken using a Nikon microscope ALPHAPHOT-2 YS2-H connected to Delta Optical DLT-CamPRO 6.3MP USB 3.0. Micrographs were collected under reflected light.

Fourier transform infrared spectroscopy

Fourier transform infrared spectroscopy (FTIR) spectra were recorded with an attenuated total reflection mode (ATR Smart Orbit Accessory, ThermoFisher Scientific) on a Nicolet 380 spectrometer (Thermo Scientific) with a diamond cell. A resolution of 4 cm^{-1} and the scanning range from 600 to 4000 cm⁻¹ were applied, and 32 scans were taken for each measurement.

Contact angle

Static water contact angle measurements were performed using the DataPhysics OCA 15EC contact angle system (DataPhysics instruments GmbH) at ambient temperature. A 2 mL drop of distilled water was used for each measurement and a photo of the drop was taken immediately after immersing the drop in the sample surface. At least five measurements were made for each composite system.

Differential scanning calorimetry

Differential scanning calorimetry (DSC) tests were performed using a Setaram Labsys TG-DTA/DSC apparatus according to EN ISO 11357-1: 2023. The samples were heated at a rate of 10°C/min in a nitrogen atmosphere at temperatures ranging from 20°C to 280°C. Important parameters of the samples, such as: glass transition temperature (T_q) , melting temperature (T_m) and enthalpy of melting (ΔH) were estimated. The percentage crystallinity for PET composites was calculated from the equation (1), according to Mondadori et al. (2011).

$$X_c = \frac{\Delta H_m}{\Delta H_m^0 \Phi} \cdot 100\% \tag{1}$$

where: X_c – crystallinity [%], ΔH_m – melting enthalpy [J/g], ΔH_m^0 – melting enthalpy of 100% theoretically crystalline PET (140 J/g) (Tariq et al., 2021), Φ -PET weight fraction in composite

Mechanical test

Tensile tests were performed according to EN ISO 527-1: 2020 using Zwick Roell 020. Furthermore, the impact test was carried out in Impact Hammer - BPI-5.5COMC - Zwick / Roell according to EN ISO 179-2:2023 (Charpy impact determination) and EN ISO 180:2023-11 (Izod impact determination). A pendulum hammer 5J was used.

Results and Discussion

As shown in Figure 2 – transparent, green PET bottles were cut and formed into composite samples using an injection moulding machine. The opacity of the samples obtained for further testing was due to

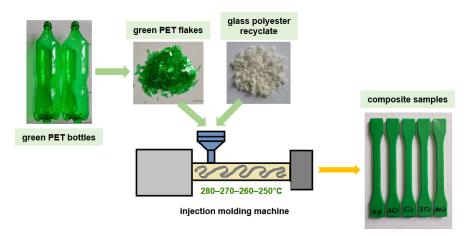


Fig. 2. Scheme of the individual stages of obtaining rPET modified with glass polyester recyclate

their thickness (the thickness of the bottle wall is 0.4 mm, while the thickness of the obtained rPET sample is 5 mm).

Some properties of rPET composites were studied after adding glass polyester recyclate in a different amount.

Surface Investigation Results

The surface of all samples appeared to be completely smooth, which is important considering the potential application of the obtained composites. If the particles of glass polyester recyclate were on the surface of these composites, there would be a risk that water would penetrate between the PET matrix and the filler. This would affect the durability of products manufactured from these materials, especially in the case of outdoor applications.

Therefore, the sample surfaces were examined under magnification using an optical microscope with reflected light.

As can be seen in the top image of Figure 3, the surface of the rPET sample was mostly smooth and even. Only the small holes were visible for rPET samples, but also for their composites. According to Monti et al. (2021), this can most likely be attributed to the formation of acetaldehyde as a degradation product that typically appears in PET-based injection-moulded components.

With an increase in the content of glass polyester recyclate on the surface of the samples, more and more filler inclusions can be seen. Their shape is diverse, from spherical and ellipsoidal to strongly elongated. The last are probably glass fibres, which are a reinforcement of polyester resin recyclate. In addition, it can be seen that there are shadows around the glass polyester recyclate particles in the rPET matrix, which may indicate incompatibility between these two materials. On the other hand, since both materials, i.e. the matrix and the filler, are polyesters, it can be expected that there may be hydrogen-type interactions between

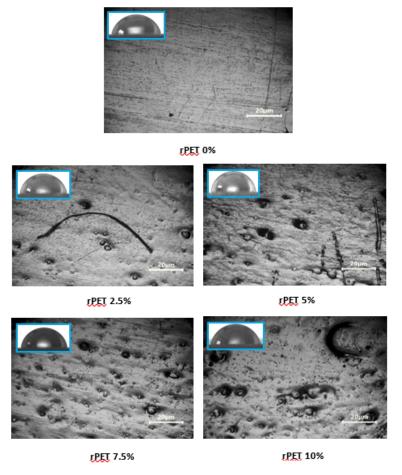


Fig. 3. Optical microscope images of the surfaces of rPET samples and their composites with 2.5%, 5%, 7.5% and 10% glass polyester recyclate additives. The upper left corner shows the shape of a water droplet applied to the sample surface during the contact angle test

them. However, considering that light in an optical microscope penetrates a certain depth into the sample, it is not possible to clearly determine whether the filler inclusions are on the surface or covered by the rPET layer. It is also difficult to determine whether the visible shadows around the added particles indicate their lack of connection with the rPET matrix or just depressions in the surface of the composite samples, which are formed in places where the filler is attached. The answer to these questions can be provided by ATR-FTIR studies and measurements of the surface water contact angle, the results of which are presented below. In addition, a scanning electron microscope examination is planned in the future, but this will be the subject of another study.

FTIR spectra analysis

FTIR with ATR is a tool that allows for identification of chemical groups located on the surface of the sample. Figure 4 shows representative spectra of glass polyester recyclate, PET flakes, rPET, and their composite with a 10% weight content of this recycled filler. For each type of the tested material, several spectra were performed, which allowed assessing the homogeneity of their chemical structure on the surface of the samples.

Peaks at 2928 cm⁻¹ and 2910 cm⁻¹ in the spectrum of the recyclate used as a filler indicate C-H stretching in aliphatic chains, implying the presence of saturated hydrocarbon segments. A distinct peak at 1716 cm⁻¹ corresponding to the carbonyl group (C=O) confirms that it is a polyester resin. Moreover, the peaks at

1599 cm⁻¹ and 1579 cm⁻¹ indicate the presence of aromatic rings, especially from the styrene monomer, providing evidence of the aromatic content in the resin. The band associated with the methyl group (CH₃) at 1376 cm⁻¹ indicates that the resin is an ortho-styrene derivative (Manikumar & Rao, 2024).

The bands of aliphatic groups on the spectra of all PET flakes, rPET, and their compounds are about 2910 cm⁻¹ and 2960 cm⁻¹. Therefore, they are slightly shifted in relation to the polyester glass filler. Much greater differences can be seen in the fingerprint area, that is: between 600 cm⁻¹ and 1800 cm⁻¹ of the spectra. It differs significantly from the resin spectra.

All the spectra performed for PET materials are identical. They show characteristic bands for this polymer corresponding to the carbonyl group (at approx. $1724 \,\mathrm{cm}^{-1}$), the oxy-ethylene groups (at $884 \text{ cm} - 1 \text{ and } 967 \text{ cm}^{-1}$), and the CH₂ in the PET ester groups of PET (at 1340 cm^{-1} and 1370 cm^{-1}) (Mecozzi & Nisini, 2019). The bands detected at 722 cm^{-1} , 871 cm^{-1} and 1407 cm^{-1} correspond to the vibrations of the CH=CH groups of the aromatic ring, while the bands at 1611 and 1504 cm^{-1} are from the C-C vibrations of this ring (Vozniak et al., 2024). Furthermore, the bands at 1090 cm⁻¹ related to the symmetric C-O stretching vibrations and the bands at $1240 \,\mathrm{cm^{-1}}$ related to the asymmetric C-O-C stretching vibrations in the ester group were identified (Królikowski et al., 2024; Hideky et al., 2024).

Some differences in the intensity of the bands at $1340~\rm cm^{-1}$ and $1370~\rm cm^{-1}$ could be observed in some spectra taken on different areas of the samples of rPET 7.5 wt.% and rPET 10 wt.% (the area marked

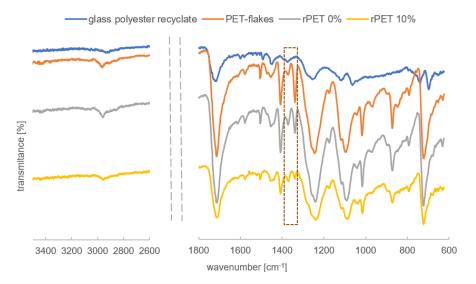


Fig. 4. Representative spectra of glass polyester recyclate, PET flakes, rPET, and their composite with a 10% recyclate content

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in Figure 4). In most cases, the intensity of the band at $1340 \,\mathrm{cm^{-1}}$ was higher than that of the band at $1370 \,\mathrm{cm^{-1}}$. However, for these two types of composites it was found that it is the other way round in several spectra. According to Mecozzi & Nisini (2019), the smaller the absorption ratio $1340 \,\mathrm{cm^{-1}}/1370 \,\mathrm{cm^{-1}}$, the more amorphous the polymer form, whereas the higher $1340 \,\mathrm{cm^{-1}}/1370 \,\mathrm{cm^{-1}}$ ratios correspond to the more crystalline structure. Therefore, although the presence of glass polyester recyclate particles on the surface of the composite samples was not detected, it should be stated that they had an influence on the chain organisation of these materials. Their presence probably disrupted the order of the rPET chains and hindered crystallite formation.

Contact angle results

The measure of wettability of a solid surface, including a polymer surface, is the value of the contact angle with a liquid, mainly water. Its value depends on several parameters, including the chemical structure of the polymer (the presence of elements in the chains that can interact with water molecules, e.g. oxygen, nitrogen), its amorphous or crystalline nature, the degree of surface uniformity, or its roughness (Benke et al., 2024; Kruszelnicka et al., 2017).

Figure 5 shows the dependence of the contact angle of the rPET surface and its composites on the amount of added glass polyester recyclate.

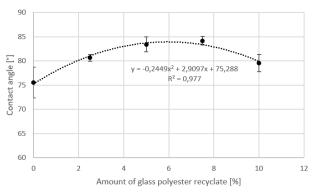


Fig. 5. Contact angle of rPET composites with a different amount of glass polyester recyclate

Considering that the potential application of the obtained composites may be the so-called small architectural products, i.e. benches, road posts, waste bins, etc., it is better for the hydrophobicity of these materials to be as high as possible. This will ensure easier run-off of raindrops, which is so-called self-cleaning, in outdoor applications and also ensure hygiene in products used in buildings. According to Prime and

Mozetič (2002) PET is a moderately hydrophobic polymer, with a static contact angle between 70 and 80°, despite the relatively large percentage of oxygen. As can be seen in Figure 5, mechanical recycling processing has no effect or only a very slight effect on the contact angle of rPET 0 wt.%, the average value of which was 75.5°.

On the other hand, the addition of glass polyester recyclate undoubtedly had an impact on an increase in the hydrophobicity of rPET. As shown above, the FTIR tests of the sample surface confirmed that the filler inclusions are located below the surface and are covered by the rPET layer. Therefore, they do not directly change the hydrophobicity of the samples. However, their presence in the rPET matrix causes a certain inhomogeneity and roughness of the surface. This means that a water droplet cannot spread easily and the wetting angle increases. When 10% by weight of recyclate was added to rPET, the sample surface became sufficiently rough for water molecules from the deposited droplet to penetrate the surface asperities, resulting in droplet spreading and a reduced contact angle, as shown in Figure 3. The wetting angle of this composite was still 4° higher than that of rPET. The highest hydrophobicity was characterised by the rPET $7.5 \text{ wt.}\% \text{ composite } (84^{\circ}).$

DSC results

To destroy the thermal memory of the sample, DSC thermograms from the second heating cycle are usually analysed. Nevertheless, we decided to interpret the results of the first heating cycle in order to see what effect post-processing in the injection process had on the thermal history of PET (Jaime-Aziare et al. 2024). The course of DSC thermograms indicates a glass transition and melting of the crystalline phase of PET. However, for composites, no thermal transformations of the glass polyester recyclate were observed because its amount in the entire mass of the sample was too small for the device to record them. Table 1 summarises the values of T_g , T_m and the values of ΔH_m of PET flakes, rPET 0 wt.% and its composites with glass polyester recyclate.

Bahramian (2001) found that the melting point of typical bottle grade PET decreased from 248°C to 236°C after processing with an extruder. As shown by the values in Table 1, the formation of PET-flakes in rPET by injection resulted in a minimal increase in the melting point, with a simultaneous slight increase in crystallinity. This discreet increase in crystallinity from 28.9 % to 29.8 % is indicated by the slight loss of transparency of the rPET flakes after drying prior to the injection moulding process.

Table 1
Thermal properties of PET-flakes and rPET composites with a different amount of glass polyester recyclate

| Sample | Glass temp. T_g [°C] | Melting temp. T_m [°C] | Enthalpy ΔH_m (J/g) | Crystallinity [%] |
|--------------------------------------|------------------------|--------------------------|-------------------------------|-------------------|
| Glass polyester recy- clate | 64.1 | 165.7 | 56.9 | _* |
| PET- flakes | 77.7 | 248.6 | 40.4 | 28.9 |
| rPET 0% | 72.3 | 251.8 | 41.7 | 29.8 |
| rPET 2.5% | 71.9 | 252.2 | 27.3 | 20.0 |
| rPET 5% | 71.7 | 254.3 | 31.2 | 23.5 |
| rPET 7.5% | 71.0 | 249.9 | 21.4 | 16.5 |
| rPET 10% | 69.2 | 252.4 | 27.8 | 22.1 |

^{*}The crystallinity of the glass polyester recyclate cannot be determined.

However, knowing that the temperature of commercial PET can vary, it is not possible to clearly state what effect the injection has on it (Awaja & Pavel, 2005). It is undoubtedly clear that the addition of glass polyester recyclate particles disturbed the arrangement of the chains, inhibiting their ordering and transformation into crystallites. Although the presence of filler particles only slightly influenced the melting temperature of the PET crystalline phase, it significantly reduced its amount. At the same time, the shape of the melting peak of the crystalline phase became narrower and sharper, indicating that the resulting crystallites became more homogeneous. The onset point values of the composites were much closer to the T_m values than in the case of rPET flakes and rPET 0%. It is expected that this lowered crystallinity will unfortunately reduce the mechanical strength of the composites compared to that of rPET, yet it may make the material less brittle.

According to the data given by Mettler-Toledo (2011), the glass temperature of virgin PET is 79.2°C, but Awaja and Pavel (2005) stated that T_g varies between 67°C and 140°C. Therefore, it is not possible to clearly determine what effect the formation of the original PET in bottles had on its glass transition temperature. However, it is known that the secondary process (obtaining samples in the injection process) reduced T_g , which should affect the elasticity of rPET.

Additionally, the addition of glass polyester recyclate lowered the glass transition temperature of rPET even more. The increased mobility of the PET chains, which acted as a plasticiser by shifting T_g toward lower values, may indicate that as a result of reprocessing they were cut into shorter chains.

Mechanical test results

The appropriate mechanical parameters of construction materials are a fundamental factor in construction. However, in the case of small architectural objects, they are only one of the elements that determine the selection of the right material. Aesthetic considerations, resistance to environmental conditions, material hygiene, and its low cost often outweigh the selection of a material with greater mechanical strength. The figures below illustrate the effect of adding glass polyester recyclate to the rPET matrix on its tensile strength (Figure 6A), the modulus of elasticity of the tensile (Figure 6B), the impact strength Charpy (Figure 6C), and the impact strength Izod (Figure 6D).

The tensile strength of the rPET samples was 40 MPa and was comparable to the strength of the primary PET, which is 40-60 MPa (Olam, 2023), slightly higher than in the Sim et al. study (2021), and at the same time slightly lower than reported by Huq et al. (2011). These decreases in the mechanical properties of rPET compared to the original poly(ethylene terephthalate) may result from the thermo-oxidative and thermomechanical degradation of the chains and their hydrolytic scission (Schyns et al., 2023).

However, contrary to expectations, the addition of polyester glass recyclate resulted in a reduction in the tensile strength of the composites compared to rPET in all the cases (Figure 6A). It is known that a significant improvement in tensile properties of the composite, resulting from the addition of a filler to the polyester matrix, is related, among others, to the increased interaction between both the components. Good interfacial adhesion is necessary to transfer the stresses from the polyester matrix to the reinforcing particles. Consequently, it improves the mechanical properties of such composites, as observed by Sim et al. (2021), who added glass fibres as a filler to rPET. As was said previously, the addition of recyclate to composites does not improve mechanical properties of the composites. This suggests that the interaction between the particles of the glass polyester recyclate does not exist or is negligible. This could confirm the observations made for the microscopic images. Meanwhile, Alshammari et al. (2022) explained such a reduction in the tensile strength of PET after adding graphite particles as a result of the agglomeration of filler particles.



The equations of the scaling curves for tensile strength and tensile modulus, depending on the glass polyester recyclate content, are represented as seconddegree polynomials. According to Alshammari et al. (2022) an increase in the tensile modulus with the increasing filler content indicates a greater degree of reinforcement and/or the inducement of a higher degree of crystallinity in the case of composites, which seems not to be confirmed by our research results. Yet, Al-Jabareen et al. (2012) stated that this increase in tensile modulus is typical for composites with inorganic fillers. In their studies of PET composites with graphite nanoparticles, Al-Jabareen et al. (2012) found the same relationship between an increase in tensile modulus and a simultaneous decrease in tensile strength as in the case of our samples. Therefore, the glass polyester recyclate particles in the rPET composites acted as an inorganic filler in the materials of Al-Jabareen et al. (2012) As can be seen in Figure 6B, the elastic modulus of the obtained composites gradually increased with an increase in the content of glass polyester recyclate particles. Although this increase was not large, it could indicate some type of interaction between the rPET matrix and the filler. However, according to Schyns et al. (2023), a small increase in elastic modulus indicates a decrease in the chain

length of rPET, which could occur during its injection in the presence of filler particles. This is confirmed by an apparent decrease in tensile strength (Fig. 6A) and a decrease in the glass transition temperature of the amorphous phase rPET (Tab. 1).

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Impact tests are dynamic tests that determine the ability of a material to withstand sudden impact loads. Theoretically, during the typical use of small architecture, such as waste bins, garden benches, and road stakes, these materials should not be subjected to such forces. However, random events such as kicking, hitting, or sitting down too quickly on a bench should be taken into account. Therefore, these materials should also have some resistance to such loads. Reprocessing of the PET waste resulted in a reduction in Charpy and Izod impact strengths. With the addition of more than 5 wt.% of glass polyester recyclate into the rPET matrix, composites have a lower impact strength (Fig. 6C). However, during the Izod impact test, the reduction of the impact strength was already observed after adding 2.5 wt.% of glass polyester recyclate (Fig. 6D). These results confirm the earlier conclusions that indicate a reduction in the length of PET chains after the secondary processing due to the injection with the addition of glass polyester recyclate.

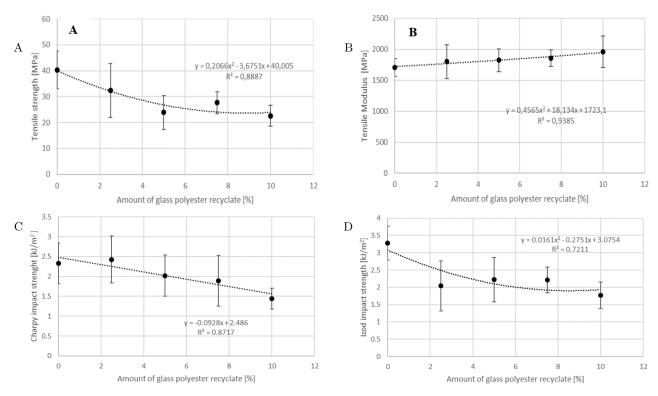


Fig. 6. Tensile strength (A), tensile modulus of elasticity (B), Charpy impact strength (C) and Izod impact strength (D) of rPET composites with different amounts of glass polyester recyclate

Conclusions

When selecting the right material for the construction of small architectural elements, various factors should be taken into account. The most important ones are: strength of the given structure taking into account the safety factor, the lowest possible cost and ease of execution, appropriate weight, ease of keeping it clean, and the appropriate aesthetics of the designed element or structure. However, considering threats related to global flooding of the natural environment with difficult-to-recycle plastic waste, all methods of the secondary use are very desirable. Of course, the presence of plastic objects in public spaces is not always aesthetically accepted, but in some situations, their durability, lack of maintenance, and ease of keeping the spaces clean outweigh the aesthetic considerations.

It is known that thermo- and oxydegradation of poly(ethylene terephthalate) chains can occur during PET reprocessing. This may result in the formation of reactive functional groups and the increased chain mobility with the reduced molecular weight (Bocz et al. 2021). When the presence of unsaturated bonds in the recycled polyester with glass fibres was considered, a reactive interaction with the rPET matrix was assumed and thus an improvement in mechanical properties of the composites without the need to add additional substances, e.g. compatibilisers.

The scope of the work included the production and testing of composites from rPET recyclate as a matrix and 2.5, 5, 7.5 and 10% by weight of glass polyester recyclate as a filler. It was found that the mechanical strength of the obtained composites is slightly lower than that of rPET alone. This indicates the lack or inactivation of unsaturated bonds in the recyclate that could react with functional groups of thermodegraded and oxydegraded rPET. However, and this is extremely important, FTIR-ATR tests indicate the absence of recyclate particles on the surface of the material, which could reduce its resistance to degradation factors. This also concerns samples with the highest, 7.5 and 10% by the weight filler content. Moreover, the obtained composites have an aesthetic appearance and their hydrophilicity is low.

To sum up, post-production glass polyester composite waste generated during the production of yachts can be a valuable secondary raw material. In combination with the coloured waste of poly(ethylene terephthalate) recyclate from post-consumer bottles it can be an interesting material to obtain small architectural objects by a simple and inexpensive mechanical method using injection moulding. What is important is that discolouration of the rPET flakes is not necessary.

In order to obtain materials with higher mechanical strength, it would be necessary to add a compatibiliser.

Thus, the research hypothesis was positively verified, and the aim of the study was achieved. The results provide a promising path to a global and cost-effective solution for the management of end-of-life glass polyester composite waste and products, which will lead to a more sustainable composite materials industry.

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