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Production of composites with added waste polyester-glass with their initial mechanical properties

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Abstract

In this article the problem of plastic recycling, and in particular waste polyester-glass, has been described. In brief, the technology for the production of new composite materials by hand, made by the contact method, using the matrix filling in the form of polyester-glass waste coming from worn-out laminates used in the construction of ship hulls. The waste used was a powdered mixture of resin particles, glass fibers, and composite particle agglomerates. A method for producing composite panels with a recyclate content of between 10% and 30% was carried out. The samples were then manufactured according to the requirements of the standard, i.e. PN-EN ISO 527-4_2000P. Samples obtained from the test plates were subjected to a static stretch test, to verify the impact of the integrated wastes on the mechanical properties of the composite. Photographs of the structure of the obtained material have also been presented. These photos showed significant differences in the composition of the resulting composites determined by the amount of waste material used. Analysis of the results indicated that increasing the amount of recyclate reduces the value of the material's strength limit and also reduces the plasticity of the material. This article has provided an introduction to more comprehensive research on the recycling of plastics.

Introduction

In Poland, about 2,000 tons of post-production waste are produced each year, and about 20,000 tons of post-consumer waste are produced in the production of fiberglass reinforced polyester products. Fiberglass reinforced polyester is used as a construction material in shipbuilding (e.g. hulls of units, superstructures), railway, automotive (e.g. car body and interior fittings, car roofs, cisterns), and aviation applications (e.g. aircraft hulls, fuel tanks, end beaks, and ballast). The widespread use of these materials in the economy has resulted in the need for the development of appropriate disposal methods for both types of waste. There are many ways to recover fiberglass from waste that can then be used as a full-fledged component, such as replacing waste for a part of the reinforcing phase in new composites (Pickering et al., 2000; Kowalska, Wielgosz & Bartczak, 2002; Pickering, 2006). Continued progress in the recycling of composites, and the need to recycle materials that have been found in the past to be unsuitable for re-use, has driven the search for new and better methods for the management of this waste (Bignozzi, Saccani & Sandrolini, 2000).

Polymer composites have been widely used due to their good chemical resistance and high mechanical strength (Jastrzębska, 2011), as well as their low density. Plastic production has increased with the GDP of countries. More sustainable composite production models and improved waste management – particularly higher recycling rates – offer great potential for savings on the cost of resources. At the same time, this would help reduce the need for transportation of raw materials as well as greenhouse gas emissions. As a result the resource savings can be significant. Plastics are almost exclusively produced from oil and now comprise 8% of the world's oil production, 4% of which is used as raw material, and 3–4% as energy for manufacturing (Hopewell, Dvorak & Kosior, 2009). According to research, recycling plastics and saving materials could have the effect of reducing the effects of climate change, reducing the need for abiotic resources and reducing the ecological toxicity of fresh water. Increasing material efficiency in the case of composites would have the greatest effect on reducing their environmental impact (EU, 2013).

In Poland, the precursor in the field of composites, especially BMC (thermosetting molding), was the Institute of Polymers of the then Szczecin University of Technology. Also in the 1990s, in collaboration with the Institut fur Werkstofftechnik of the University of Kassel, research was carried out on the recycling of such materials. These works concerned the possibility of using various recycled fractions from SMC in polyester moldings, related to technological limitations, modification of recipes, flow of molds with recycled forms, etc. The possibilities of using comminuted polyester-glass laminates in lightweight and polymer concrete were also investigated, Solvolysis of hardened polyester resin (Gorący, 2006; Błędzki, Gorący & Urbaniak, 2012).

Also in Japan, for many years, researchers have been looking for methods of recycling composites, based mainly on polyester and epoxy resins, reinforced with carbon fiber. They researched various methods of recycling raw material, e.g. by glycolysis of cured polyester resins, supercritical water vapor and water degradation, and dissolving the material in strong solvents. Work continues, but there are no systematic solutions for the recycling of composites in this country (Kanemasa, 2011). Research in this field has also been carried out in Poland from the year 2000. It is also worth mentioning the work carried out at the Technical University of Opole (Rutecka, Śleziona & Myalski, 2004; Rutecka, Kozioł & Myalski, 2006), and numerous inquiries recently initiated by the industry, and also work done at the Maritime University in Gdynia (Jastrzębska, 2011; Jastrzębska & Jurczak, 2011).

The Silesian Technical University (Rutecka, Kozioł & Śleziona, 2005) has undertaken research into the use of composite waste as a polymer matrix filler in composite materials. Waste materials produced during the production of composite constructions manufactured by the "WENTECH" company in Imielin were used to study the influence of the addition of composite waste on the properties of laminates. The waste used for the tests was: post-production waste, defective and damaged products, and used composite products. The matrix in these composites was Estromal Polyester Resin 14.LM-01, reinforced with mat or fiberglass fabric. The reinforcement volume was approximately 30%. The crusher used was the SK 100 cross-bevel mill manufactured by Retch. In order to determine the particle size, a sieve analysis was performed. It showed that the recyclate was a powder fraction with a particle size of less than 0.3 mm. Furthermore, it should be noted that the polyester-glass recycled was a heterogeneous material consisting of a mixture of polyester resin flakes and very fine glass fibers, or agglomerates of composite particles (Gawdzińska et al., 2017).

Methodology of preparation of test samples

A fragment of polyester-glass scrap was obtained from the original composite material of the hull of a ship which had been manufactured in Poland in the 1980s. It was a technological breakthrough when cutting openings for new deck equipment. This scrap was initially crushed by a hammer and then crushed in a specially prepared plastic waste processing plant (the so-called crusher). After crumbling, the waste was put through a sieve with a mesh diameter of 1.2 mm to obtain a recyclate that served as filler which was then added to the composite matrix. Recyclable granulation was ≤ 1.2 mm. The preparation of a recyclate with such granulation is a complex process and strongly affects the environment – a large amount of highly irritating dust was produced. Therefore, the preparation of the test material required appropriate safety equipment and security. A concept was developed in the laboratory of the Department of Fundamental Techniques of the Faculty of Mechanical Maritime University in Gdynia, and a plastic crushing plant was developed.

Figure 1. A station for the processing of composite materials obtained by the Maritime University in Gdynia

Figure 2. Hardened composite panels: 1 – without recyclate, 2 – 10% recyclate, 3 – 30% recyclate (photo taken with 13 Mpixel camera, Sony IMX 214 sensor and focal length 27 millimeters)

The next step after obtaining a recyclate with the appropriate granulation was the choice of how to prepare the composite panels. It was decided to use manual lamination by the contact method, using the mat as reinforcement (Strong, 1989), and as a matrix for all the following materials, epoxy resin, orthophthalic rigid POLIMAL 109A of "Organika-Sarzyna" S.A. with hardener and accelerator.

Figure 1 shows a station for the processing of composite materials obtained by the Maritime University in Gdynia.

Firstly, a "clean composite" plate with 0% recyclate content was produced from a combination of the above resin and glass mat, which was further used as reference material. This plate consisted of 12 layers of resin-impregnated glass mat with added hardener and accelerator. The weight percent of the glass mat in this plate was 36%. Next a composite panel of 10 wt% recyclate content of granularity ≤ 1.2 mm was produced. This plate was made of 10 layers of glass mat which made up 26% by weight. The pre-prepared waste was swapped for a part of the mass of the mat in relation to the mass of the reference plate. The warp accounted for 64% of the weight of the plate. The last layer was a composite panel with 30% recyclate content of granularity ≤ 1.2 mm. This plate was made of 2 layers of glass mat which is 6% by weight. The pre-prepared waste was swapped for a part of the mass of the mat in relation to the mass of the reference plate. The matrix was 64% by weight. Figure 2 shows the finished, hardened composite panels before samples were cut from it.

Methodology of research

To determine the mechanical properties of composites with added polyester-glass waste, samples needed to be prepared in accordance with the

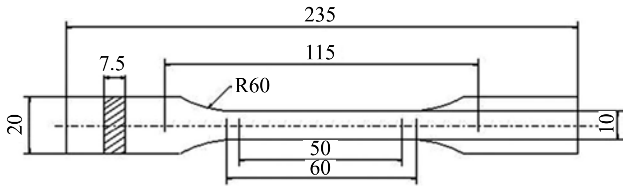

Figure 3. Dimensions of samples prepared for static stretching, according to standard PN-EN ISO 527-4_2000P

requirements of the applicable standard PN-EN ISO 527-4_2000P. The dimensions of the samples are shown in Figure 3. Composite samples were made by water jet cutting. This method ensured that the dimensions of each sample were maintained. Figure 4 shows the prepared samples ready for testing on the endurance machine.

Figure 4. Samples prepared for static stretching: in the foreground a sample of 30% waste, then a sample group with 10% recyclate and clear samples without recyclate

The strength tests of the samples, prepared as described above, were performed on the MPMD P10B hydraulic power transmission system using TestXpert II, software version 3.61, written by Zwick & Roell (Figure 5) with the use of the extensometer Epsilon Model 3542.

Figure 5. The universal strength machine from Zwick Roell

Results of the research

In order to determine the effect of recyclate proportions on the mechanical properties of the composite, the samples were subjected to a static stretch test on the endurance machine shown above. During each test the samples were elongated using an extensometer for axial measurements. Figure 6 shows the graphs of σ–ε for samples with a recyclate content of 0, 10, and 30%.

Due to the fact that the composite material was made with the recyclate content coming from composite scrap that was actually over 30 years old, that formed part of the hull of an existing watercraft manufactured at the Gdynia Maritime Academy, it is a completely new material, especially since it was hand made by the contact method. In the first phase of the study, it was decided to check the reproducibility of the results obtained for a series of randomly selected samples from each composite board, i.e. 0% recyclate material (reference material) and plates with 10% and 30% waste added as matrix filler, and replacing the weight share of the glass mat in relation to the share of the mat in the reference material.

By analyzing the above results, Table 1, and the graphs of the static tensile test – Figure 6 for each group of samples, it can be concluded that the method selected for the production of the above-described composite panels and the mere performance of the samples ensures the reproducibility of the results for the sample, regardless of where the sample was taken from on the plate surface. At the same time it was noted that increasing the amount of recyclate has a significant effect in reducing the mechanical properties of the composite. The Young's modulus (E_t) decreased as the amount of waste in the sample increased. Increasing the amount of recyclate reduced the strength limit of the composite. In case of elongation of the samples (ε_m), as the amount of recyclate increased, the elongation at destruction decreased. After adding more recycled material, the composite became much more brittle.

The reduction in the strength of the materials tested, depending on the amount of waste included, strongly correlates with the increased porosity of the composite structure. The mixture of recyclate with the matrix and the matting of the glass mat caused the recyclate particles to be located between the layers of the fabric, thus having a direct effect on the properties of the composite. The reinforced glass mat was less efficiently saturated compared to the

Table 1. Summary of the results of the static composite tensile test with the addition of recyclate

	$L_{0\,\mathrm{Trav}}$	L_0	E_t	σ_m	ε_m	b	h
	mm	mm	MPa	MPa	%	mm	mm
1 – 0% recyclate	128.4204	50	7748.747	105.5553	1.79235	10.05	7.55
2 – 0% recyclate	132.0818	50	7348.42	115.3876	2.142618	9.95	7.04
3 – 0% recyclate	134.5791	50	8008.947	105.8981	1.732728	10.03	7.04
4 – 10% recyclate	126.8868	50	7102.685	83.80287	1.828101	9.84	7.34
5 – 10% recyclate	128.7527	50	7668.565	83.02501	1.759299	9.8	6.63
6 – 10% recyclate	130.7406	50	6549.639	86.79197	2.189828	9.88	7.75
7 – 30% recyclate	129.6582	50	4449.016	22.87513	0.757042	9.99	6.83
8 – 30% recyclate	128.3377	50	3821.872	24.59061	0.87887	9.98	8.42
9 – 30 % recyclate	127.0878	50	4020.72	28.46265	0.928936	9.97	7.85

$L_{0\,\mathrm{Trav}}$ – inital length (traverse), L_0 – initial length (extensometer), E_t – Young's modulus, σ_m – tensile strength limit, ε_m – total elongation, b – sample thickness, h – height.

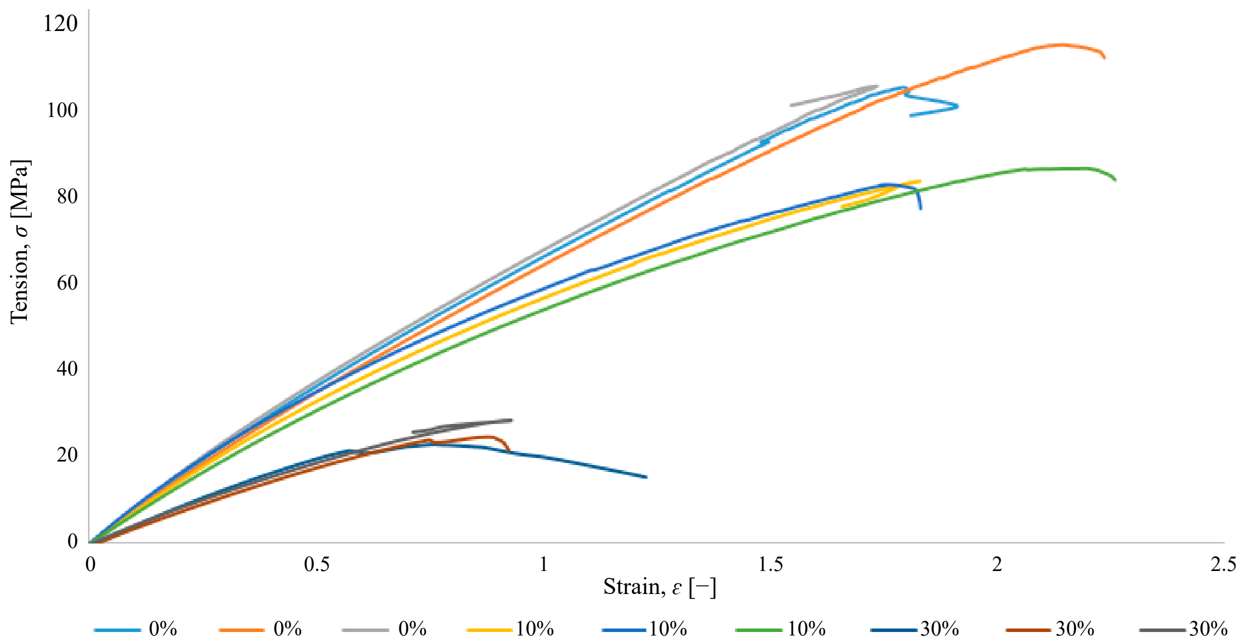

Figure 6. Static stretch test for samples with recyclate content of 0, 10, and 30%, with recyclate granulation ≤ 1.2 mm

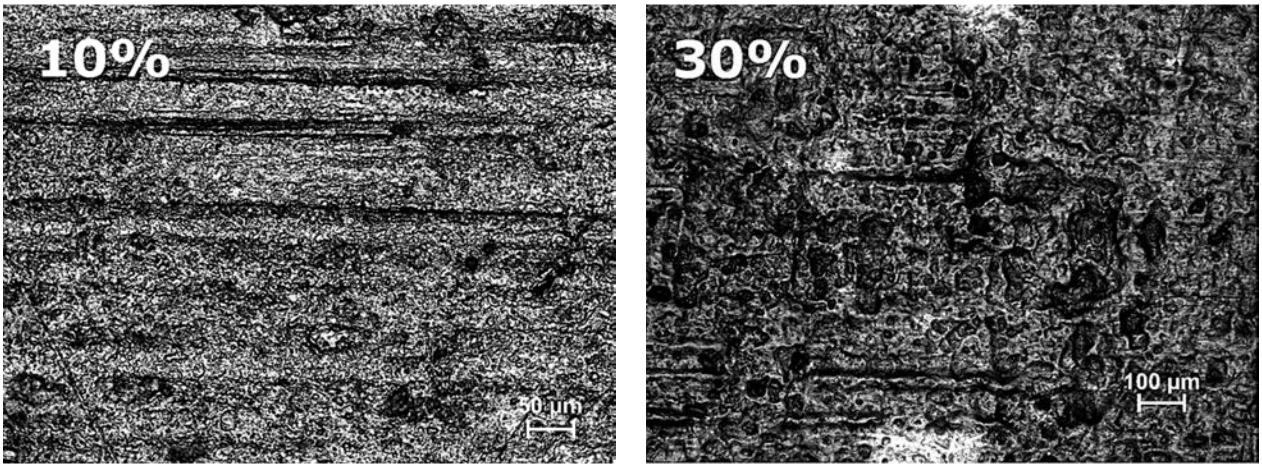

Figure7. Cross section of the composite with 10% and 30% filler

non-recyclable composite. In addition, introducing waste into the structure can have an effect on the formation of airspace inside the material (pores), as shown in Figure 7, which further lowers the strength of these composites.

Conclusions

The scrap polyester-glass hull of the vessel was crushed and put through a sieve with a mesh diameter of 1.2 mm. The resulting waste was not subjected to any other mechanical, thermal, or chemical treatment. It was therefore a powder fraction consisting of glass fiber resin particles and composite particle agglomerates. The recyclate was added to the epoxy, orthophthalic, rigid epoxy resin as the matrix fill, in the amount of weight replacing the reinforcement portion (mats) with the amount of reinforcement used in the reference material (pure composite) respectively:

- with a sample of 10% recyclate (reducing the proportion of mats from 36% to 26%);
- with a 30% recyclate sample (reducing the proportion of mats from 36% to 6%);

which reduced the strength of plastics to the level of:

- 77.6% of the reference material's strength;
- 23.2% of the reference material's strength.

In addition, material with a 30% recycled content has an accordingly reduced percentage elongation to break average of 1.89% for the reference material (pure composite without recyclate).

The results obtained from the tensile strength test showed that the recyclate content of the material has a significant influence on the mechanical properties of the composite. From the point of view of static loads (static stretching test), it is preferable to use about 10% added waste in the new composite. Increasing the proportion of waste in the laminate mass to 30% significantly lowers the strength of the material and also reduces its plasticity.

Results for samples with a recyclate content of 10% (with a decrease in the strength limit of 22.4%, relative to the strength of the pure composite), showed no apparent change in the elongation of the samples relative to the reference material. This indicates the need for further research seeking the optimum percentage of recyclate in the total mass of the finished composite. The ideal percentage of added waste would maximize waste management whilst not significantly reducing the durability and plasticity of the material.

It was initially shown that the addition of recyclate, in significant quantities i.e. 30%, had a negative effect on the structure of the material. Increasing the amount of waste increased the number and the extent of internal imperfections in the composites. The glass mat was resealed with a mixture of resin and recyclate with the introduction of filler particles between the fibers of the mat. These particles caused the formation of airspaces (pores), which significantly weakened the material and reduced the strength of the composite and made it less plastic. In addition, a recyclate of 30% by weight in the composite matrix significantly limited the technological ability of the matting. The matrix became very dense and the manufacturing of composites using the manual contact method became significantly impeded – at the boundary of the material in general. A further increase in the fraction of powder fraction in the composite appears to be unfeasible at this stage of the research.

For the purpose of this article Preliminary studies were aimed at presenting the technology for producing recycled composite material, as well as the verification of the reproducibility of the results of the test, as well as the influence the amount of polyester-glass waste used had on the mechanical properties of the resulting composites. Initial test results were satisfactory in terms of material technology and reproducibility of the strength test results of the samples produced. Continued advances in the recycling of composites, and materials that have been deemed unsuitable for reuse, are encouraging researchers to find new opportunities for waste utilization. The global problem, however, remains unresolved and further research and a wider scope is needed to find out how to reuse polyester-glass waste. Without improved product design methods and better waste management measures, the amount of plastic waste in the EU will continue to grow as production increases.

Plastics that have been found in the environment – particularly in the marine environment – can survive there for hundreds of years. Plastics are not inert. Traditional plastics contain a large number, and sometimes a high percentage, of chemical additives that may impair the functioning of the human endocrine system, be carcinogenic or possess other toxic effects. These substances may, in principle, leach into the environment in varying amounts. Particular concerns have been raised about small and very fine particles (the so-called micro-plastic) formed by decades of photodegradation and mechanical abrasion. Plastic microchips and chemical additives may enter the marine fauna's digestive tracts, and therefore there is a serious risk that they might contaminate the food chain. Inadequate land-based management, and in particular the negligible recovery rates of plastic waste, deepen the problem of marine pollution by plastic materials, which is now one of the most important new global environmental issues (EU, 2013).

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