

The impact of polyester-glass recyclate on the hardness and structure of composites

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Abstract

The attractive characteristics of polyester-glass composites have led to their use in many industries, but using them as structural elements requires knowing their mechanical properties. This paper presents processing methods of polyester and glass scrap and their use in the production of new composites. This scrap, called the recyclate, was pre-crushed, ground, and then passed through sieves to obtain the desired fractions. Composite materials with added recyclate were made by hand lamination. Composites were made and then used to conduct appropriate tests to determine the degree of cure of the resin, and to determine the effect of the polyester-glass recyclate content on the hardness of composites. For this purpose, rectangular panels with a thickness of $g = 8$ mm were produced by manual lamination. Each plate contained 0% glass mat, 10% resin, and 20% recyclate with granulation ≤ 1.2 mm and ≤ 3 mm. Then, test samples were formed from each plate. Hardness measurements were performed using the ball-pressing method. In addition, structural studies were carried out to determine the correlation between the structural and mechanical properties of the discussed materials. The obtained test results showed that the recyclate content and its granulation clearly affected the mechanical properties of the tested composite materials.

Introduction

Plastics are used in almost all industries (Królikowski, 2012), and their wide application has impacted the emergence of new technologies to produce these materials, aimed at automating and accelerating their production processes. They are included in many types of composites, with the matrix materials of epoxy resins, polyesters, etc. They have also been used as reinforcement materials in mats and fabrics, etc. The basic factors affecting the hardness of composites are (Naplocha & Samsonowicz, 2001) their fiber properties, fiber content, warp type, fiber-warp fusion quality, stress state, and the density of crystal lattice defects around fibers (Jiang, et al., 1994; Kang & Yun, 1996). The first step in obtaining a composite is the selection of an appropriate fiber material. Composites with fiber

reinforcements currently dominate the market for composite materials due to their superior mechanical strength and low weight. Often, the use of more expensive fibers with much better properties allows a maximum strengthening effect to be obtained. Unfortunately, this is associated with higher costs, which prevents their use. The fibers used for the production of composites may be continuous (elementary) or discontinuous (staple fibers, whiskers) (Rajczyk & Stachecki, 2011). When selecting a reinforcement, it is also possible to use numerous products made of single fibers: roving, mats, fabrics, pre-impregnations, and shaped elements (Oczóś, 2008). The second important factor affecting the hardness of composites is the type of matrix (Sobczak, et al., 1995).

As technologies related to the use of polymer composites (Gawdzińska, et al., 2017) have

developed to use fiber reinforcements, the amount of post-consumer waste increases, which has increased interest in the use of these waste materials (Rutecka, Ślezionek & Myalski, 2004; Rutecka et al., 2005). One important aspect is an appropriate means of processing these materials because the form in which the material is processed will affect its subsequent use (Habaj, 2008). There are many methods to recover glass fibers from waste, so that they can then be used as full-value components (Asokan, Osmani & Price, 2009), replacing part of the reinforcement phase in new composites (Kowalska, Wielgosz & Bartczak, 2002; Pickering, 2016). Continuous progress has been made in developing methods to recycle composites and materials (Bignozzi, Sacconi & Sandrolini, 2000). Until recently, many of these materials were considered unfit for reuse, which has prompted an investigation into developing new, superior methods to reuse these materials (Jastrzębska, 2011; Jastrzębska & Jurczak, 2011; Błędzki, Gorący & Urbaniak, 2012; Garbacz & Kyzioł, 2017; Panasiuk & Hajdukiewicz, 2017; Kyzioł, Panasiuk & Hajdukiewicz, 2018).

This article presents the influence of recycled (recycled scrap) polyester and glass on the properties of layered composites subjected to hardness tests. In addition, characteristics of the structures of the tested materials were determined, and their impact on hardness was determined.

Test sample preparation methods

The analyzed materials were composites with added polyester and glass recyclate. The recyclate was sourced from a fragment of a ship's hull produced in Poland in the 1980s. The hull fragment was initially crushed by a hammer and then crumbled with a crusher. After being milled and passed through a series of sieves, the material was divided into relevant fractions and used to produce the composite materials, which were made by hand laminating. The recyclate was used in the following quantities: 0%, 10%, 20% with granulations ≤ 1.2 mm and ≤ 3 mm. A detailed description of the production of composites with different recycled content is presented in previous works (Panasiuk & Hajdukiewicz, 2017; Kyzioł, Panasiuk & Hajdukiewicz, 2018; Panasiuk, 2018).

Table 1 shows the percentage of components that make up polyester-glass composites with added recyclate. The following symbols are used to simplify composite identification:

- LR0 – composite without recyclate;

Table 1. The content of composite materials made by hand laminating

No.	Symbol	Number of mat layers	%resin content	% mat contents	% recycled content
1	LR0	12	66%	34%	0%
2	LR10.1.2	10	64%	26%	10%
3	LR10.3.0	10	62%	27%	10%
4	LR20.1.2	3	69%	11%	20%
5	LR20.3.0	3	70%	10%	20%

- LR10.1.2 – composite with 10% recyclate content with granulation ≤ 1.2 mm;
- LR10.3.0 – composite with 10% recyclate content with granulation ≤ 3.0 mm;
- LR20.1.2 – composite with 20% recyclate content with granulation ≤ 1.2 mm;
- LR20.3.0 – composite with 20% recyclate content with granulation ≤ 3.0 mm.

The next stage was to prepare samples for testing and investigate the hardness of composite materials based on the presence of recyclate.

Research methods

The hardness of samples was measured by pressing a ball in accordance with the PN-EN ISO 2039-1: 2004 standard (Plastics – Determination of hardness – Part 1: The method of pressing the ball). A Qness Q250M universal hardness tester equipped with an indenter for testing plastics was used in all tests. In addition, Qpix T12 software was used to prepare a measurement report and fully archive the measurement data, including the recorded images displaying the imprint.

The method involved pressing the ball under a given load into the surface of the tested fitting and then measuring the depth of the impression under the load. The area of the ball imprint was calculated from the depth of the fingerprint. Hardness (HB) is defined as the quotient of the ball indenter of the hardness tester to the surface of the impression surface caused by the indenter ball after a specified load duration. The ball diameter was 5 ± 0.05 mm. The measurement process consisted of applying a pre-load $F_0 = 9.8 \pm 0.1$ N, at a minimum distance of 10 mm from the edge of the fitting. Next, a sensor used to measure the depth of the impression was set to zero, and a load was applied so that the depth of the impression was in the range of 0.15–0.35 mm. The test time was 30 s. To determine the hardness of the materials, 10 measurements were performed on samples of each material.

Figure 1 presents a diagram of the hardness measurement points on samples.

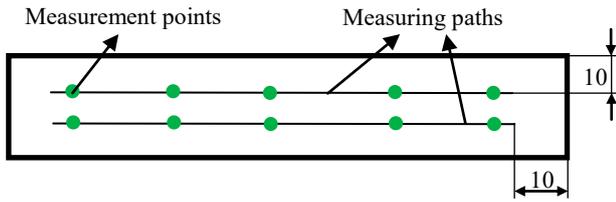


Figure 1. Diagram of hardness measurement points on the sample

Figure 2 shows an example of a ball impression on a sample LR10.1.2 (10-fold magnification). Structural investigations were carried out on an Axiovert 25 optical microscope with 50× to 500× magnification.

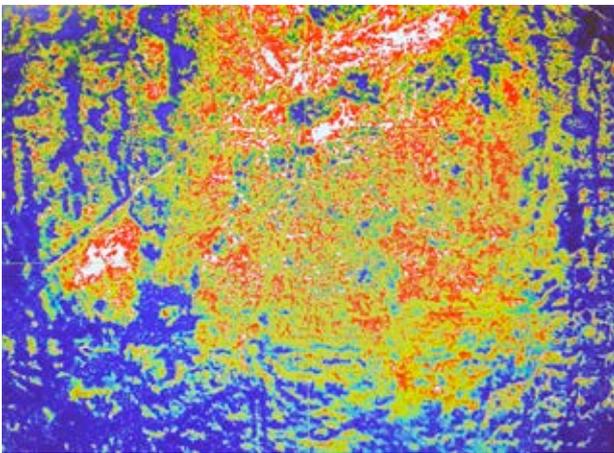


Figure 2. Sample printout LR10.1.2 (10×) (Panasiuk, 2019)

The aim of the study was to measure the amount of recycle, its distribution, and size of granules in the structure because these quantities significantly impact the mechanical properties of the tested materials. Tests were carried out on cut pieces from each composite. The cross-sectional areas were prepared using abrasive papers with gradations of 320, 800, and 1200, and then polished with a diamond polishing slurry with a grain size of 3 μm.

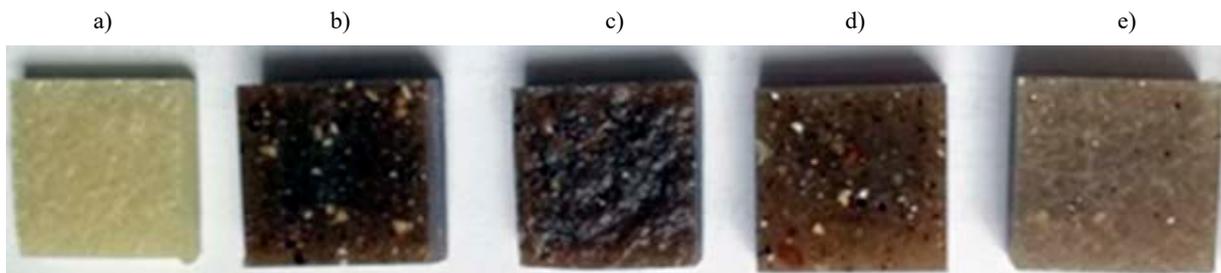


Figure 3. Samples prepared for optical microscopy observation: a) LR0, b) LR20.3.0, c) LR20.1.2, d) LR10.3.0, e) LR10.1.2 (Panasiuk, 2019)

Figure 3 presents images of prepared samples from composite materials for optical microscopy observations. Structure observations were made of the cross-sections of samples. Primary observations included the amount of air pores in the composite and the adhesion of the recycled particles. The assessment of material structures will allow for the proper analysis of the mechanical dependence of the tested materials on their structural properties.

Results

Three hardness tests were performed with each composite, and 10 measurements were taken on each sample. The average results of hardness tests are presented in Table 2.

Table 2. Hardness measurement results obtained by ball pressing composites made via manual lamination (Panasiuk, 2019)

Material	HB	Standard deviation	ΔHB
	N/mm ²	N/mm ²	%
LR0	161	17.28	0
LR10.1.2	140	13.96	13.0
LR10.3.0	134	19.4	16.8
LR20.1.2	94	16.85	41.6
LR20.3.0	96	9.38	40.4

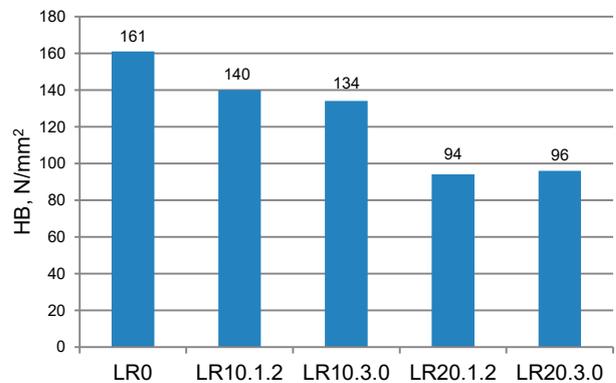


Figure 4. Influence of recycle content and granulation on hardness (based on (Panasiuk, 2019))

Figure 4 presents a bar graph illustrating the impact of the recyclate content and its granulation on the hardness of composites made by manual lamination. The hardness test results presented in Table 2 and Figure 3 show that the highest hardness of the composite without recyclate was 161 MPa. The addition of 10% recyclate reduced the hardness by about 15%, and the addition of 20% recyclate reduced the hardness by more than 40% compared with the composite without recyclate. Hardness tests complement mechanical tests (strength, impact resistance, and bending) and have shown that increasing the recycled content decreases the mechanical properties of composite materials. The impact of granulate in the range of 1.2 mm to 3 mm is negligible (Garbacz & Kyzioł, 2017; Panasiuk & Hajdukiewicz, 2017; Kyzioł, Panasiuk & Hajdukiewicz, 2018).

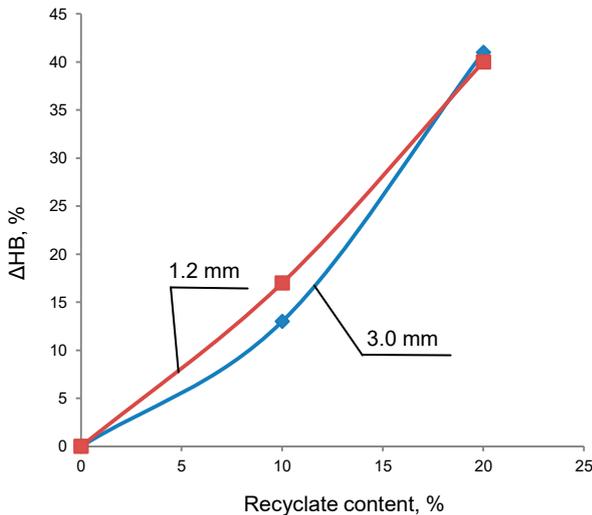


Figure 5. The impact of recycled content and granulation on composite hardness (Panasiuk, 2019)

Figure 5 shows that the granulate slightly affects the composite hardness.

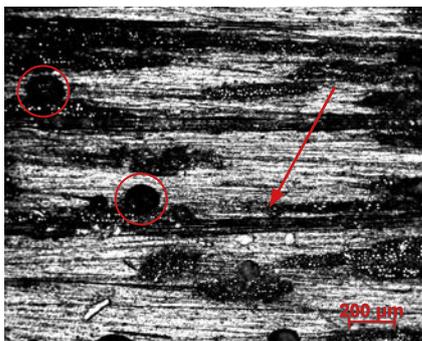


Figure 6. Structure of composite without recyclate made by manual lamination – LR0 (50x) (Panasiuk, 2019)

Figure 6 shows that the structure of a non-recyclate composite (LR0) made by manual lamination contains many air pores, especially when reinforcing. An important aspect is the adhesion between the resin and the fibers. In this material, the boundary between the fibers and the resin is visible (marked with an arrow).

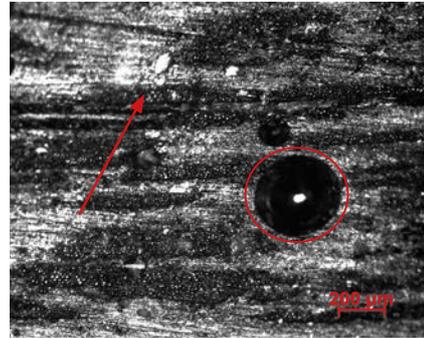


Figure 7. Composite structure with 10% recycled content, granulate size ≤ 1.2 mm, made by manual lamination – LR10.1.2 (magnification 50 \times) (Panasiuk, 2019)

Figure 7 shows the structure of a composite with 10% recyclate and a granulation ≤ 1.2 mm (LR10.1.2). The image shows that the structure of this composite is clearly different from the structure of the composite without recyclate. In the case of LR10.1.2, both air pores (marked in the image in a circle) and recyclate particles are visible (marked with an arrow).

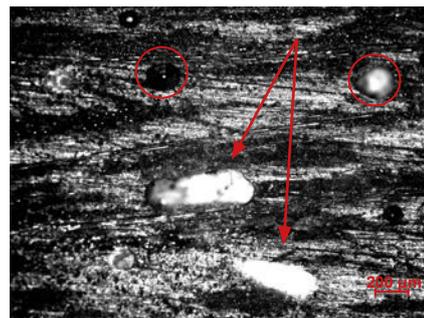


Figure 8. The structure of the composite with a 10% recyclate content, a granulate size of 3.0 mm, made by manual lamination – LR10.3 (magnification 50 \times) (Panasiuk, 2019)

Figure 8 presents an image the structure of a composite with 10% recyclate and a granulation ≤ 3.0 mm (LR10.3). Large recyclate granulates are clearly visible, which weaken the bond between the reinforcement and the resin (marked with an arrow). In addition, there are also air pores which emerged at the border with the reinforcement.

Conclusions

The results obtained here have made it possible to draw final conclusions regarding the impact of recycle content and granulate size on the hardness of polyester-glass composites.

1. The addition of polyester-glass recycle reduces the hardness of the composite.
2. The addition of 10% recycle reduces the hardness of the composite by about 15%, and the addition of 20% recycle reduces the hardness by more than 40% compared with the composite without recycle. When granulate size ranged from 1.2 mm to 3 mm, the hardness of the polymer was not substantially affected.
3. The results of the hardness tests of composites are consistent with the results of tensile strength, bending strength, and impact strength tests. Combined, the results showed that increasing the polymer content decreased the mechanical properties of the composite.
4. However, a higher recycle content in the composite decreased the mechanical properties of the produced composite, but larger amounts of recycle led to a non-workable composite material.
5. The decrease in the mechanical properties with an increase in the recycled content was caused by a reduction in the amount of reinforcement in the material, which reduced the adhesion of composite components. In addition, structure studies showed that the way in which the components were included in the composite without recycle match the plastic material. In the case of composites with added recycle, fragile cracks were observed at the turn point. Structural observations have also shown that there are clusters in which the amount of recycle in the composite significantly reduces the mechanical properties of the material.

References

1. ASOKAN, P., OSMANI, M. & PRICE, A.D.F. (2009) Assessing the recycling potential of glass fibre reinforced plastic waste in concrete and cement composites. *Journal of Cleaner Production* 17, 9, pp. 821–829.
2. BIGNOZZI, M.C., SACCANI, A. & SANDROLINI, F. (2000) New polymer mortars containing polymeric wastes. Part 1. Microstructure and mechanical properties. *Composites Part A: Applied Science and Manufacturing* 31, 2, pp. 97–106.
3. BŁĘDZKI, A.K., GORĄCY, K. & URBANIAK, M. (2012) Możliwości recyklingu i utylizacji materiałów polimerowych i wyrobów kompozytowych. *Polimery* 9, pp. 620–626.
4. GARBACZ, G. & KYZIOŁ, L. (2017) Application of metric entropy for results interpretation of composite materials mechanical tests. *Advances in Material Science* 17, 1 (51), pp. 70–81.
5. GAWDZIŃSKA, K., SZYMAŃSKI, P., BRYLL, K., PAWŁOWSKA, P. & PIJANOWSKI, M. (2017) Flexural strength of hybrid epoxy composites with carbon fiber. *Composites Theory and Practice* 17, 1, pp. 47–50.
6. HABAJ, W. (2008) Technologia kompozytów polimerowych wzmacnianych krótkim włóknem aramidowym wykonanych metodą RTM. *Problemy Techniki Uzbrojenia* 37, 105, pp. 61–73.
7. JASTRZĘBSKA, M. & JURCZAK, W. (2011) Modyfikacja kompozytów z odpadami poliestrowo-szklanymi. *Przetwórstwo Tworzyw* 17, 6, pp. 460–463.
8. JASTRZĘBSKA, M. (2011) Wpływ nanonapełniacza NanoBentu® ZW1 na właściwości kompozytów z recyklatem poliestrowo-szklanym. *Kompozyty* 11, 2, pp. 111–113.
9. JIANG, J.-Q., LIU, H.-N., MA, A.-P. & TAN, R.-S. (1994) The structure and tensile properties of Al-Si alloy hybrid reinforced with alumina-aluminosilicate short fibre. *Journal of Materials Science* 29, 14, pp. 3767–3773.
10. KANG, C.G. & YUN, K.S. (1996) Fabrication of metal-matrix composites by the die-casting technique and the evaluation of their mechanical properties. *Journal of Materials Processing Technology* 62, 1–3, pp. 116–123.
11. KOWALSKA, E., WIELGOSZ, Z. & BARTCZAK, T. (2002) Utylizacja odpadów laminatów poliestrowo-szklanych. *Polimery* 47(2), pp. 110–116.
12. KRÓLIKOWSKI, W. (2012) *Polimerowe kompozyty konstrukcyjne*. Warszawa: PWN.
13. KYZIOŁ, L., PANASIUK, K. & HAJDUKIEWICZ, G. (2018) The influence of granulation and content of polyester-glass waste on properties of composites. *Journal of KONES*, 4(25), pp. 223–229.
14. NAPLOCHA, K. & SAMSONOWICZ, Z. (2001) Twardość materiałów kompozytowych o zwiększonej zawartości włókien. *Kompozyty (Composites)* 1, 2, pp. 199–202.
15. OCZOŁ, K.E. (2008) Kompozyty włókniste – właściwości, zastosowanie, obróbka ubytkowa. *Mechanik* 7, pp. 579–592.
16. PANASIUK, K. & HAJDUKIEWICZ, G. (2017) Production of composites with added waste polyester-glass with their initial mechanical properties. *Scientific Journals of the Maritime University of Szczecin, Zeszyty Naukowe Akademii Morskiej w Szczecinie* 52(124), pp. 30–36.
17. PANASIUK, K. (2018) Analysis of technologies for producing composites with polyester-glass recycle. *Scientific Journal of Polish Naval Academy* 3(214), pp. 63–73.
18. PANASIUK, K. (2019) *Analiza właściwości mechanicznych kompozytów warstwowych z recyklatem poliestrowo-szklanym*. PhD thesis, Gdynia: UMG.
19. PICKERING, S. (2016) Recycling technologies for thermoset composite materials – current status. *Composites Part A: Applied Science and Manufacturing* 8, pp. 1206–1215.
20. RAJCZYK, M. & STACHECKI, B. (2011) Współczesne materiały kompozytowe wybrane kierunki rozwoju nowych technologii. In: Bobka, T. & Rajczyk, Z. (Eds) *Budownictwo o zoptymalizowanym potencjale energetycznym*. Częstochowa: Wydawnictwo Politechniki Częstochowskiej, pp. 202–211.
21. RUTECKA, M., KOZIOŁ, M., MYALSKI, J. & ŚLEZIONA, J. (2005) Wykorzystanie odpadów kompozytowych jako wypełniacza osnowy polimerowej w materiałach kompozytowych. *Kompozyty (Composites)* 5, 2, pp. 68–73.
22. RUTECKA, M., ŚLEZIONA, J. & MYALSKI, J. (2004) Estimation of possibility of using polyester-glass fiber recycle in laminates production. *Kompozyty (Composites)* 9, pp. 56–60.
23. SOB CZAK, N., SOB CZAK, J., KARAMARA, A. & DYB CZAK, S. (1995) *Squeeze Casting of Non-ferrous Metal Matrix Composites Reinforced with Carbon Felt*. International Conference “Cast Composites’95”, Zakopane, 18–20.10.1995, pp. 87–90.