

В.П. Кашицький^[0000-0003-2346-912X], О.Л. Садова^[0000-0002-6152-5447], А.В. Чернов, Ю.І. Крась*Луцький національний технічний університет***ФОРМУВАННЯ ЕПОКСИКОМПОЗИТІВ НА ОСНОВІ РЕЦИКЛОВАНИХ
НАПОВНЮВАЧІВ СИНТЕТИЧНОГО ПОХОДЖЕННЯ**

Представлено методику переробки скловолокнистих полімеркомполімерних матеріалів методом механічного подрібнення з наступною класифікацією частинок та формуванням епоксикомполімерних зразків. Визначено підвищення фізико-механічних характеристик епоксикомполімерних матеріалів на основі вторинної сировини, що структуровані за кінцевої температури 170°C. Встановлено зниження міцності на стискання та вмісту гель-фракції епоксикомполімерного матеріалу в результаті введення до складу композиції наповнювача в кількості більше 200 мас.ч., однак використання наповнювача в кількості 600 мас.ч. забезпечує зростання ударної в'язкості епоксикомполімерів. Використання частинок вторинної сировини меншого розміру покращує технологічність композиції та міцність на стиснення епоксикомполімерів.

Ключові слова: епоксидне в'язуче, фракція, термічна обробка, міцність на стиснення, гель-фракція, ударна в'язкість, характер руйнування.

V. Kashytskyi, O. Sadova, A. Chernov, Yu. Kras

**FORMATION OF EPOXY COMPOSITES BASED ON RECYCLED FILLERS OF
SYNTHETIC ORIGIN**

A method of processing glass-fiber polymer composite materials by mechanical grinding with subsequent classification of particles and formation of epoxy composite samples is presented. An increase in the physical and mechanical characteristics of epoxy composite materials based on secondary raw materials, structured at a final temperature of 170°C, is determined. A decrease in the compressive strength and gel fraction content of the epoxy composite material as a result of introducing a filler into the composition in an amount of more than 200 wt. parts is established. However, the use of a filler in an amount of 600 wt. parts provides an increase in the impact strength of epoxy composites. The using of smaller particles of secondary raw materials improves processability of the composition and increases the compressive strength of epoxy composites.

Key words: epoxy binder, fraction, heat treatment, compressive strength, gel fraction, impact strength, fracture behavior.

Formulation of the problem. Over the past few decades, polymer composites have replaced many metal or ceramic materials in various applications due to their ease of processing, corrosion resistance, light weight and cost-effectiveness. Such materials provide an optimal ratio between weight and mechanical properties, which is practically unattainable for many traditional materials, so their introduction into the aviation, automotive industries and the construction of large structures is quite relevant. The demand for carbon and glass fibers is constantly increasing, since such fibers are almost entirely used as reinforcement in plastic matrix composites (mainly epoxy type). The consumption of a huge amount of synthetic polymer materials and fibers in composites creates a serious problem for recycling and waste management. Most high-performance polymer composites are based on thermosetting polymer materials, which are not recyclable, since their internal three-dimensional structure with covalent bonds creates obstacles to the decomposition and recycling of used products. Therefore, there is a need to conduct research that will determine the technology for processing composites based on thermosetting materials, which is of great importance for resource conservation and environmental protection.

Analysis of recent research and publications. Glass fiber reinforced polymer composites (GFRP) are polymer composites that are widely used in electronics, electrical engineering, construction, automotive, shipbuilding and aviation industries due to their high specific strength and corrosion resistance [1]. The requirements of sustainable development and the principles of circular economy require the development of technologies for processing polymer composites based on thermosetting polymers, which usually contain a significant amount of inorganic filler. Forming processing of polymer composite material is very difficult because of the heterogeneous nature of the polymer composite material. Therefore, thermosetting polymers do not melt or dissolve, and their processing is difficult [2]. In most cases, glass fiber polymers end up in solid waste landfills [3] or incinerators, which leads to a negative impact on the environment.

According to current research, scientists recommend several alternative technologies for GFRP recovery, including mechanical, chemical and thermal methods [4–6]. Mechanical processing involves mechanical reduction of the size of composite waste by grinding [7]. The starting material is reduced by impact and shear to a size that allows the fragments to pass through a specified size of the screen holes of the shredder. Typically, recyclates are in the form of flakes, fiber-rich and resin-rich fractions [8].

© В.П. Кашицький, О.Л. Садова, А.В. Чернов, Ю.І. Крась

Chemical processing can include solvolysis, hydrolysis and acid cleavage using solvents, water, glycols and acids. In this case, the process ensures the dissolution of the polymer matrix to separate it from the filler. The thermal processing process involves high temperatures and the use of complex equipment, which, together with chemical treatment, limits the widespread use of such processing methods.

Mechanical and physical methods are most often used due to the simplicity of the equipment design. However, the disadvantage of the mechanical method is that the fillers cannot be obtained in their original form, since during grinding the fillers lose their strength and technological properties. In addition, it is difficult to remove the structured polymer from the filler surface.

In [9], positive results of mechanical processing of fiberglass waste were noted, since primary oil-based resources are saved. Mechanical processing remains an effective option for fiberglass processing [10], especially from an environmental point of view, since the use of a closed loop and secondary raw materials allows for additional environmental benefits. In addition, it was found that the processes of recovering products from glass fiber polymer composites (GFRP) waste require significantly less energy (0.17–1.93 MJ/kg) compared to the production of primary fiberglass (13–54 MJ/kg) [11]. Recycled fiberglass fragments are usually in the form of short fibers or powder, unlike primary, long fibers, which affects the technology for forming products from recycled materials.

Setting tasks. The aim of the work is to study the influence of the fractional composition of fragments from the processing of glass-fiber polymer composites on the mechanical characteristics of recycled epoxy composites formed at different heat treatment temperatures.

Presentation of the main material. The secondary raw material is fragments of electronic boards based on fiberglass, which were dried at a temperature of 110° C for 2 hours. Next, additional grinding of the fragments was carried out, after which the powder was sieved using sieves with a hole size of 0.7 mm and 0.5 mm. The resulting powder was mixed with epoxy binder to a homogeneous mass and placed in the mold matrix. The composition was compressed using a hydraulic press and left for structuring for 24 hours at room temperature. Heat treatment of the samples was carried out in a stepwise mode: 1 hour at a temperature of 50° C, 1 hour at a temperature of 100° C, 5 hours at a temperature of 170° C. The highest compressive strength is achieved by epoxy composite materials with a content of finely dispersed filler (fraction less than 0.5 mm) of 150 wt. parts per 100 wt. parts. epoxy binder under the condition of heat treatment for 3 h at a final temperature of 150° C (Fig. 1). Compressive strength of biocomposites decrease by 6–8% with an increase in the final temperature of heat treatment 170° C, which is associated with the appearance of residual stresses due to excessive compression of the composition during the molding process under the influence of static load, since fine-dispersed fillers have a high ability to compact compared to fillers of a larger fraction.

The compressive strength of biocomposites decrease to 280 MPa after heat treatment at a final temperature of 150° C in the case of using a fine-dispersed filler in an amount of 200 wt. parts. This is due to the increased content of the filler, which has high surface energy and requires an optimal amount in the ratio of system components. Increasing the final heat treatment temperature to 170°C leads to an increase of 17–18% in the compressive strength compared to 150°C, since a higher heat treatment temperature provides a higher degree of structuring due to the formation of additional chemical bonds between the components of the epoxy polymer system.

Biocomposite materials contained with 600 wt. parts of the filler have less compressive strength by 68–70% after heat treatment at a temperature of 150°C compared to epoxy composites containing the filler in an amount of 150 wt. parts. This is due to the excess content of filler, which is not wetted by the epoxy binder. With an increase in heat treatment temperatures to 170°C, epoxy composites obtain a high strength value (292 MPa), which indicates the dominant effect of heat treatment on the structuring processes, since the mobility of the segments of the epoxy matrix macromolecules increases.

The ultimate strength of epoxy composite materials containing larger particles (more than 0.5 mm) is 356 MPa (Fig. 2), which is almost the same as the strength of epoxy composites (360 MPa) containing fine particles less than 0.5 mm in size. With an increase in the heat treatment temperature to 170° C, there is a slight increase in the compressive strength to 363 MPa, which is due to the ability of the epoxy matrix macromolecules to perform oscillatory movements in the structure, which is characterized by a less dense arrangement of filler particles compared to the compact arrangement of particles in epoxy composites containing a fine fraction (less than 0.5 mm)

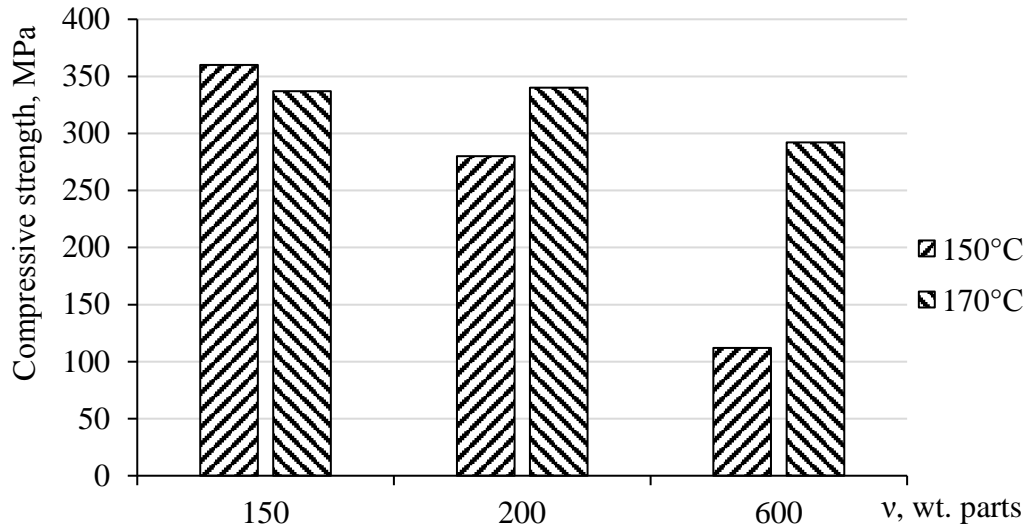


Fig. 1. The influence of the content of powder particles of secondary raw material of a fraction less than 0.5 mm on the compressive strength of epoxy composites

Increasing the filler content to 200 wt. parts leads to a decrease in the compressive strength, since larger particles are more chaotic arrangement and they are difficult to compactly arrange in the volume of the epoxy polymer matrix. Increasing the temperature to 170° C provides a slight increase in compressive strength due to the possibility of forming a larger number of chemical bonds.

Compressive strength of biocomposites decrease to 95 MPa in the case of containing of a filler in an amount of 600 wt. parts, which is due to insufficient thermal effect at a temperature of 150° C of heat treatment. Increasing the temperature to 170° C provides a compressive strength limit in the range of 280–290 MPa, since additional structuring of the epoxy composite system occurs.

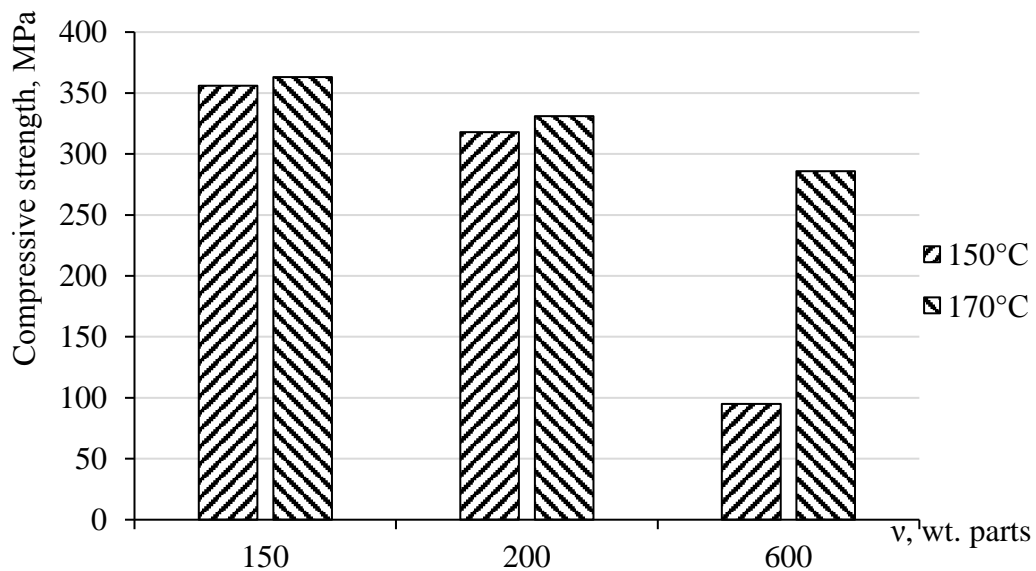


Fig. 2. The influence of the content of powder particles of secondary raw of a fraction larger than 0.5 mm on the compressive strength of epoxy composites

The gel fraction content in the epoxy composite material with a filler content of 150 wt. parts, which contains particles of a fraction with a size to 0.5 mm, is 88% in the case of sample processing at a final temperature of 150° C (Fig. 3). With an increase in the heat treatment temperature to 170° C, the degree of structuring increases to 92%, which is associated with an increase in the rate of chemical reaction and the formation of a larger number of chemical bonds.

Gel fraction content of biocomposites decreases slightly with an increase in the filler content to 200 wt. parts, since the density of the epoxy composite material decreases due to the introduction of larger particles. The structuring of the system can be improved by using the temperature of heat treatment of 170° C. This is possible due to increased mobility of the segments of the epoxy matrix macromolecules.

The content of the gel fraction is the lowest in the case of forming a material with a filler content of 600 wt. parts. This is due to the presence in the system of a large number of particles that are poorly wetted by the binder.

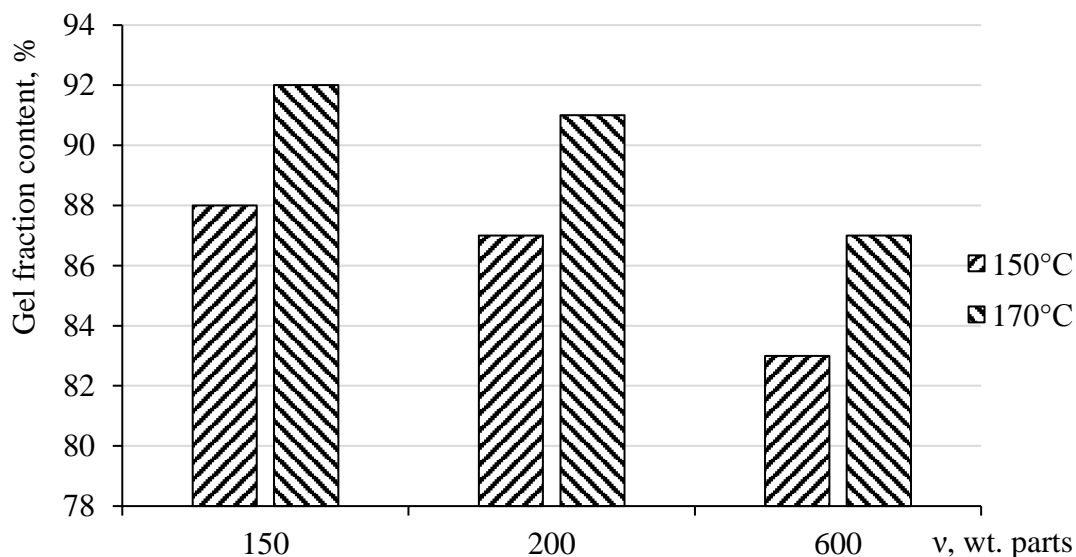


Fig. 3. The influence of the content of powder particles of secondary raw material of a fraction less than 0.5 mm on the content of the gel fraction of epoxy composites

The impact strength of epoxy composites with a filler content of 150 wt. parts, which are structured at a final heat treatment temperature of 150° C, is 3.2 kJ/m² (Fig. 4). With an increase in the heat treatment temperature to 170° C, the investigated characteristic increases to 4.5 MPa, which is associated with an increase in the material's resistance to dynamic loads due to better structuring of the epoxy polymer matrix.

Increasing the filler content to 200 wt. parts leads to a decrease in the impact strength of the material in the case of a heat treatment temperature of 150° C, which is explained by the insufficient thermal effect on the formation of a highly filled system. With an increase in temperature to 170° C, the impact strength increases to 4.8 kJ/m² due to the possibility of forming additional chemical bonds.

The temperature of 150°C is insufficient for the formation of epoxy composites with a filler content of 600 wt. parts, since the ability of the binder to wet the surface of the filler particles is reduced, however, after processing at a temperature of 170°C, the impact strength increases to its maximum value of 5.7 kJ/m². The increase in this characteristic occurs by 20–22% compared to the impact strength of epoxy composites containing 150 wt. parts of the filler. Such an increase in impact strength is due to the presence of a significant number of obstacles to crack propagation under the influence of dynamic loading in a rigid epoxy polymer matrix. Particles larger than 0.5 mm in size form obstacles in the epoxy polymer matrix that are able to dissipate kinetic energy better than smaller filler particles.

The structure of epoxy composites depends on the size of the filler particles based on secondary raw materials and its content in the epoxy polymer matrix. A dense structure of epoxy composites is formed in the case of using a filler in an amount of 150 wt. parts, since the particles are fully wetted by the epoxy polymer binder (Fig. 5, a). With an increase in the filler content to 200 wt. parts, small pores are present on the surface of the epoxy composite sample (Fig. 3.5, b). This indicates about an insufficient binder content in the system. Craters and pores are present on the surface of the epoxy composite material, which contains 600 wt. parts of filler (Fig. 3.5, c), which indicates a low degree of compaction of the epoxy composite material due to poor wettability.

Under the influence of static loading, the epoxy composite samples are destroyed with the formation of a main crack, which indicates the formation of a rigid structure of the epoxy polymer matrix.

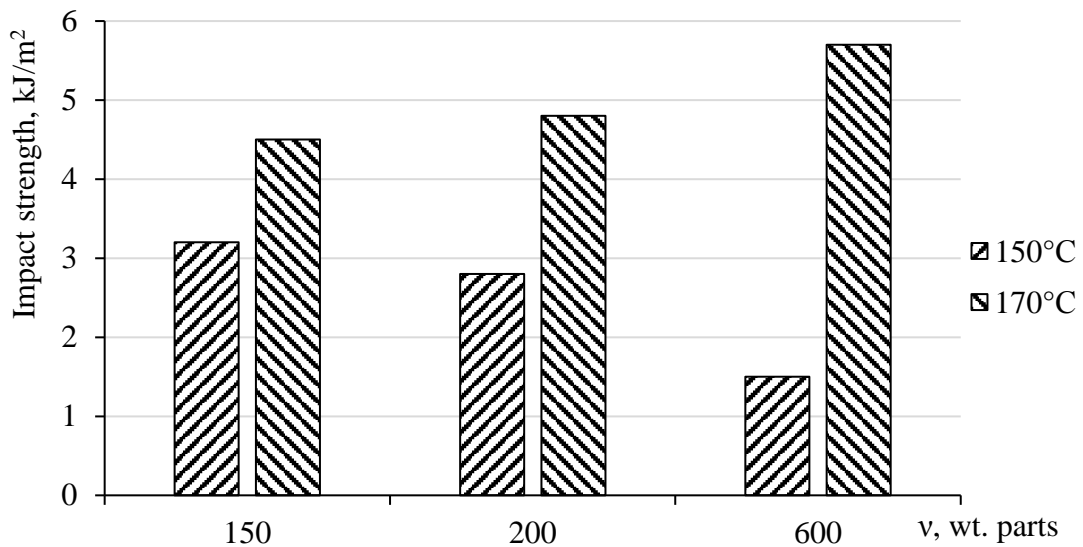
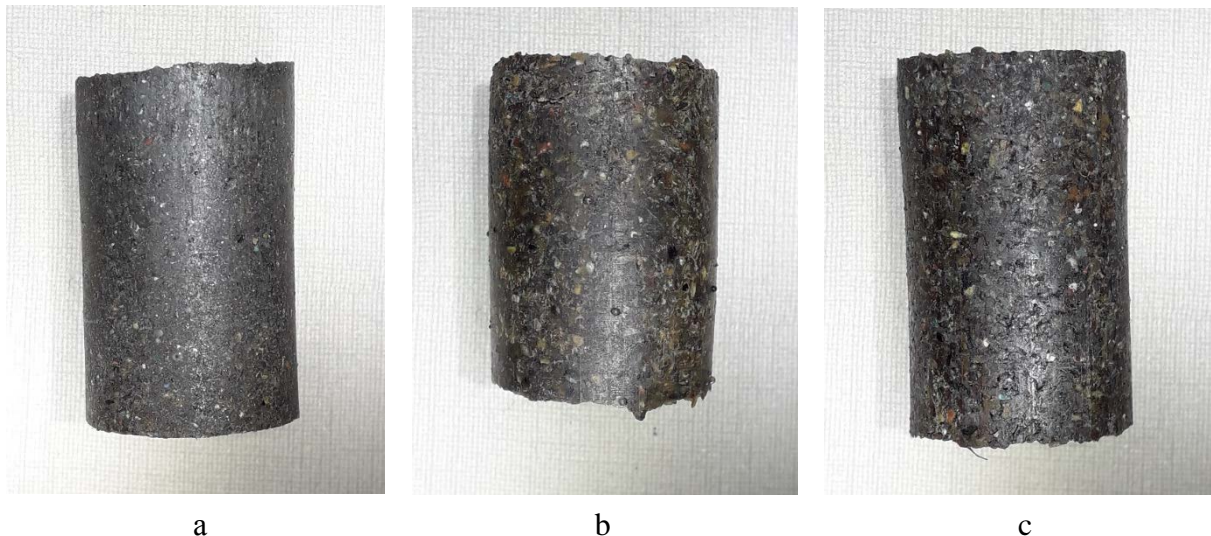


Fig. 4. The influence of the content of powder particles of secondary raw material of a fraction less than 0.5 mm on the impact strength of epoxy composites

The main crack in the epoxy composite sample with a particle fraction of less than 0.5 mm and a filler content of 150 wt. parts is located at an angle of 75° (Fig. 6, a). In the case of compression of epoxy composite samples with a filler content of 200 wt. parts, the main crack propagates at an angle of 65° (Fig. 6, b).



This indicates the presence of plastic deformation, since the sample is subjected to tangential loads. In this case, the resistance of the epoxy composite material to the influence of static loading decreases. Epoxy composite samples containing 150 wt. parts of the filler particles larger than 0.5 mm fail with the formation of a main crack located at an angle of 50° (Fig. 7, a). In the case of failure of epoxy composite samples with a filler content of 200 wt. parts, the main crack is located at an angle of 45° (Fig. 7, b). This indicates an increase in the influence of tangential loads, which cause plastic deformation, since such a material has a reduced resistance to static loads.

Conclusions and prospects for further research. Epoxy composite materials with a content of finely dispersed filler (fraction less than 0.5 mm) have the highest compressive strength (360 MPa) when using a filler in an amount of 150 wt. parts, which is associated with a uniform and compact distribution of filler particles.

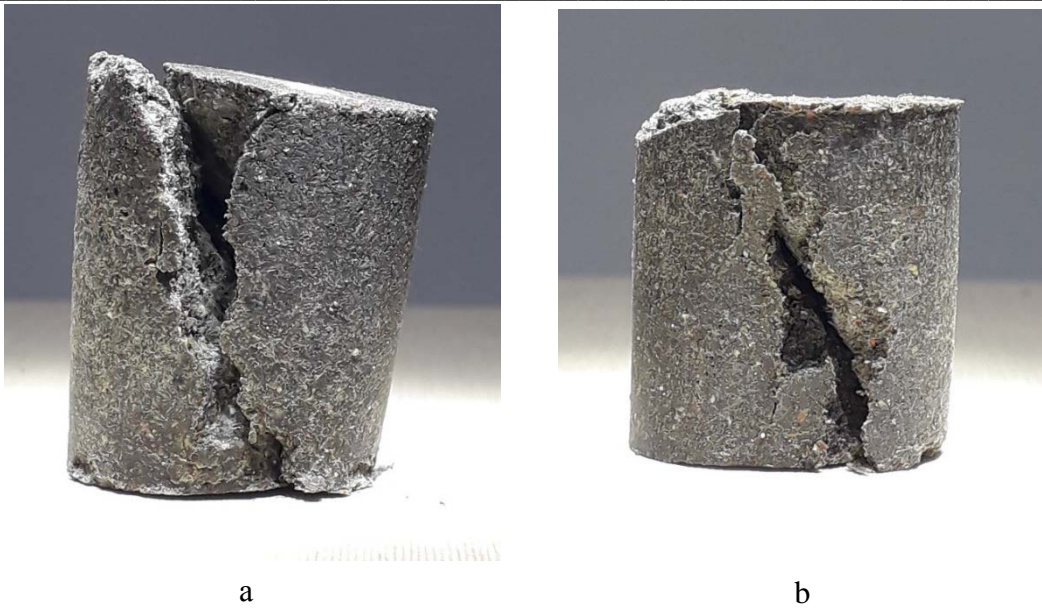


Fig. 6. General appearance of epoxy composite samples after compression with filler content (fraction less than 0.5 mm): a – 150 wt. parts; b – 200 wt. parts.



Fig. 7. General appearance of epoxy composite samples after compression with filler content (fraction more than 0.5 mm): a – 150 wt. parts; b – 200 wt. parts.

With an increase in the temperature of heat treatment to 170° C, the degree of structuring increases to 92%, which is associated with an increase in the rate of chemical reaction and the formation of a larger number of chemical bonds.

The impact strength of epoxy composites increases to its maximum value of 5.7 kJ/m² when epoxy composites are treated at a temperature of 170° C and a filler content of 600 wt. parts, since particles larger than 0.5 mm form obstacles in the epoxy polymer matrix that are able to dissipate kinetic energy better than smaller filler particles.

The compressive strength of biocomposites decrease with increasing particle size and their content in the epoxy polymer matrix. That fact can be explained by the irregular arrangement of filler particles in the volume of the epoxy composite material. In the future, it is planned to carry out a combined treatment

of fiberglass fragments with the additional use of thermal or chemical methods in combination with a mechanical treatment method in order to increase the degree of purification of fibers from the polymer matrix.

References

1. Giorgini L., Leonardi C., Mazzocchetti L., Zattini G., Cavazzoni M., Montanari I., Tosi C., Benelli T. (2016). Pyrolysis of fiberglass/polyester composites: Recovery and characterization of obtained products. *FME Transactions*, 44(4), 405–414.
2. Pickering S. J.: Recycling technology for thermoset composite materials-current status, *Composites Part A*, Vol. 37, pp. 1206-1215, 2006.
3. Conroy A., Halliwell S., Reynolds T. (2006). Composite recycling in the construction industry. *Composites Part A: Applied Science and Manufacturing*, 37(9), 1216–1222.
4. Castro M. A. C., Carvalho J. P., Ribeiro M. C. S., Meixedo J. P., Silva F. J. G., Fiúza A., Dinis M. L. (2014). An integrated recycling approach for GFRP pultrusion wastes: Recycling and reuse assessment into new composite materials using fuzzy Boolean nets. *Journal of Cleaner Production*, 66, 420–430.
5. Neri E., Berti B., Passarini F., Vassura I., Giorgini L., Zattini G., et al. (2018). Application of LCA methodology in the assessment of a pyrolysis process for tyres recycling. *Environmental Engineering and Management Journal*, 17(10), 2437–2445.
6. Silva R. V., Oliveira M. P., Souza F. G. (2020). Recycling of fiberglass reinforced plastics: A review. *Materials Research*, 22(Suppl. 1), e20190389.
7. Oliveux G., Dandy L. O., Leeke G. A. (2015). Current status of recycling of fibre reinforced polymers: Review of technologies, reuse and resulting properties. *Progress in Materials Science*, 72, 61–99.
8. Shuaib N. A., Mativenga P. T. (2016). Effect of process parameters on mechanical recycling of glass fibre thermoset composites. *Procedia CIRP*, 48, 134–139.
9. Vladimirov V., Bica I. (2017). Mechanical recycling: Solutions for glass fibre reinforced composites. *Proceedings of the International Symposium “The Environment and the Industry” (SIMI 2017)*.
10. Vladimirov V., Bica I. (2019). Methodology and calculation model for recycling of composite construction products. *E3S Web of Conferences*, 85, 07016.
11. Shuaib N. A., Mativenga P. T. (2016). Energy demand in mechanical recycling of glass fibre reinforced thermoset plastic composites. *Journal of Cleaner Production*, 120, 198–206.

Дата надходження статті до видання: 05.03.2026

Дата прийняття статті до друку після рецензування: 24.03.2026

Дата оприлюднення 14.04.2026