

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

Луцький національний технічний університет

ВПЛИВ ТЕРМІЧНОЇ ОБРОБКИ НА ФОРМУВАННЯ ЕПОКСИКОМПОЗИТНИХ МАТЕРІАЛІВ ДЛЯ БАЛІСТИЧНОГО ЗАХИСТУ

Досліджено стійкість епоксикомпозитних матеріалів з вмістом склотканини до впливу статичних та динамічних навантажень. Визначено оптимальний режим термічної обробки епоксиолімерних склопластиків, яка забезпечує високу адгезійну міцність полімерної матриці до наповнювача, а також високу ударну в'язкість та міцність епоксикомпозитів. Досліджено характер руйнування епоксиолімерних склопластиків з вмістом арамідної тканини та склотканини під впливом динамічного навантаження ударника каліброваного розміру. Визначено вплив природи наповнювача та щільності плетіння склотканини на стійкість до крихкого руйнування склопластиків за оптимального режиму термічної обробки. Розроблені епоксикомпозитні матеріали з підвищеною стійкістю до динамічних навантажень доцільно використовувати для виготовлення елементів індивідуального балістичного захисту.

Ключові слова: склотканина; епоксиолімерна матриця; тривалість обробки; структурування; адгезія; ударна міцність.

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

INFLUENCE OF HEAT TREATMENT ON THE FORMATION OF EPOXY COMPOSITE MATERIALS FOR BALLISTIC PROTECTION

The resistance of epoxy composite materials containing glass fabric to static and dynamic loads was investigated. The optimal heat treatment regime for epoxy fiberglass materials was determined. This ensures high adhesion strength of the polymer matrix to the filler. It also provides high impact toughness and strength for the composites. The nature of the destruction of the epoxy fiberglass materials containing aramid fabric and glass fabric under the influence of dynamic loading of a calibrated size impactor was investigated. The influence of the nature of the filler and the density of the glass fabric on the resistance to brittle fracture of the fiberglass materials under the optimal heat treatment regime was determined. The developed epoxy composite materials with increased resistance to dynamic loads are expedient to use for the manufacture of elements of individual ballistic protection.

Keywords: glass fabric; epoxy polymer matrix; processing duration; structuring; adhesion; impact strength.

Formulation of the problem. Synthetic fiber-reinforced polymer composites are often used because of their high processability, specific strength, corrosion resistance and low cost. Due to their high strength-to-weight and rigidity-to-weight ratios, fiberglass has a wide range of applications in the aerospace, marine, automotive and chemical industries, and in the manufacture of household and sporting goods. Fiberglass has a relatively low density and can also be adapted to the influence of various factors by changing the sequence of laying the fiber filler to provide high strength and stiffness in the direction of high load.

The specified properties of fiberglass depend on such factors as the matrix and fiber material, their volume fractions, fiber orientation, stress distribution and strain rate, loading conditions and the nature of the bonds at the fiber-polymer interface. The reaction at the interface of the polymer matrix and fiber plays an important role in determining the overall mechanical characteristics. It provides an effective way of transferring stresses to the fibers through the matrix. Recently, there has been a significant increase in the large-scale production of fibers and epoxy composite materials reinforced with glass fiber, which requires research into the influence of epoxy composite formation modes on their mechanical characteristics.

Analysis of recent research and publications. A polymer composite material consists of two or more different materials, which allows obtaining properties that are much better than the properties of the individual initial components. In this case, it is possible to combine different materials, but it is necessary to take into account the condition of ensuring macroscopic homogeneity of the structure of the polymer composite material and microscopic homogeneity of the polymer matrix. Modern polymer composite materials are usually optimized to achieve a certain balance of properties in a certain range of applications under the influence of electrical, thermal and environmental factors [1].

The unique characteristics of each phase affect the mechanical properties of fiberglass [2, 3]. The mechanical properties of the polymer matrix affect the efficiency of load transfer between fibers, which further affects the mechanical properties of fiberglass. In addition, the smooth surface of glass fibers (GF), which have a small number of active functional groups, ensures the formation of a structure with an optimal ratio of the size of glass fibers and the polymer matrix. The mechanical properties of fiberglass are mainly improved by the use of high-performance fibers and increased adhesion in the polymer-fiber system. In spite of the fact that glass fiber (GF) has low tensile strength compared to other reinforcing fibers such as carbon, basalt and aramid, it provides more economic advantages. The mechanical properties of fiberglass also depend on the performance of the polymer matrix and the compatibility between the fibers and the polymer matrix.

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Glass fibers are one of the most versatile industrial materials, as they are produced from raw materials that are currently available in constant supply [4]. They have useful properties: resistance to any chemical attack, hardness, stability, transparency and inertness. In addition, they also have good mechanical properties of the fiber: rigidity, flexibility and strength. Epoxy resin is often used as a matrix, which is a type of reactive polymer containing epoxy groups. Such groups are able to react with themselves or with many reagents, for example, phenols, amines, thiols in the presence of a catalyst [5]. Epoxy polymers have a diverse range of applications in industry for various purposes, as they usually have higher mechanical properties and greater thermal and chemical stability compared to any other type of resin.

The properties and mechanical behavior of a fiber-reinforced composite mainly depend on the strength of the fiber and its modulus of elasticity. They also depend on the chemical stability of the composite, the strength of its matrix, and the interface that acts between the fiber and the matrix for reliable stress transfer [6]. The mechanical behavior of reinforced composites depends on the strength of the fibers and the Young's modulus, stability, and strength of the matrix [7]. The appropriate orientation and composition of the glass fibers provides the desired characteristics and functional properties of fiberglass, which correspond to the rigidity and density of aluminum.

The increase in the mechanical properties of polymer composites occurs due to the interweaving of fibers, although microbending modes are activated, which reduces the compressive strength [8]. Strengthening the polymer matrix reduces delamination; however, it often decreases the modulus, a critical factor in enhancing the strength of polymer composites. Modified resins are usually used in pre-impregnated materials to resist delamination. However, the high cost of prepreg materials often limits their use in large-scale polymer composite production. Therefore, inexpensive composite materials such as glass fiber are sought to create such products. Glass fiber, polymers and carbon fiber have good mechanical properties, but in the recent period of development their use has been limited to some technological applications. Glass fibers have relatively higher strength and rigidity and low cost. Therefore, in recent developments, glass fibers are mainly used as a reinforcing material for the production of lightweight and relatively strong composite materials.

The mechanical properties of fiberglass are lower than those of polymer composites reinforced with carbon and aramid fibers, so methods for improving the mechanical properties of fiberglass are constantly being studied. In [9], an optimized processing method based on multiphase structures for fiberglass was developed, which significantly improves their tensile strength. The results show an increase of 10.64% in the specific load of fiber glass bundles due to improved bond between fibers. Combinations of factors, such as layer thickness and resin chemical composition, significantly affect the tensile ability and failure mode of fiber glass. Fiberglass with four layers of glass fabrics contribute to more efficient load transfer across the matrix-fiber interface, which provides optimal tensile properties for fiberglass. Resin binders with more reactive functional groups (-OH) strengthen the fiber-resin bond, which further improves the tensile properties of fiberglass. Nanomaterials and functional hardeners are used to improve the multiphase structure of fiberglass.

The glass fiber composites developed in [10] have six layers of glass fiber fabric with a density of 270 g/m² and two-component epoxy resin system. Tests on composites with the smallest fiber fraction revealed a higher bending stress value (464.8 MPa) compared to those with a larger fiber fraction (415.5 MPa). The failure mechanisms included delamination, crack propagation between fibers, fiber rupture and complete failure of the composite. The absorbed specific energy has similar values for composites with different contents of glass fabric layers. This can be explained by the fact that increasing the thickness of the composite can lead to a larger number of local defects and uneven distribution of components that accumulate during the manufacture of the composite.

Setting tasks. Determination of the optimal temperature and duration of exposure in the thermal field for epoxy fiberglass materials with increased resistance to dynamic loads.

Presentation of the main material. The prepared epoxy composition was applied to the steel plate matrix. After that, a reinforcing component impregnated with an epoxy binder was applied. The operation was repeated until the completion of the impregnation stage of all layers of reinforcing filler and the next compression of the workpiece on a hydraulic press with a force of 5 kN. At the first stage, structuring of epoxy composites was carried out at a room temperature of 20±2°C for 24-26 hours. In order to improve the mechanical characteristics, heat treatment was carried out in a step mode in a drying cabinet of the SNOL75/400 model with an initial temperature of 50°C and a final temperature of 150-170°C (Table 1). The mechanical processing of epoxy composite plates consisted in removing the remnants of the epoxy polymer and obtaining samples of the correct shape.

Table 1 – Forming modes of epoxy composite plates

Heat treatment				
1 mode	2 mode	3 mode	4 mode	5 mode
50° C – 1 hour, 100° C – 1 hour, 150° C – 3 hour	50° C – 1 hour, 100° C – 1 hour, 150° C – 5 hour	50° C – 1 hour, 100° C – 1 hour, 170° C – 3 hour	50° C – 1 hour, 100° C – 1 hour, 170° C – 5 hour	50° C – 1 hour, 100° C – 1 hour, 190° C – 3 hour

The adhesive strength of epoxy polymers formed according to mode No. 1 heat treatment is 18 MPa (Fig. 1). There is a slight increase in adhesive strength to 18.3 MPa in the case of an increase in exposure at the second stage of heat treatment to 2 hours at a temperature of 100°C. This is due to an increase in the mobility of segments of epoxy binder macromolecules at elevated temperature, which leads to the formation of additional chemical bonds between the end groups of epoxy macromolecules.

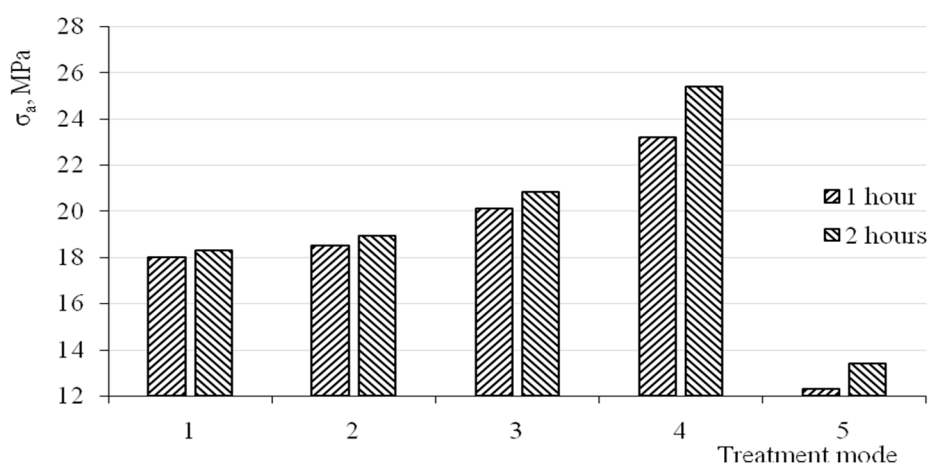


Fig. 1. Adhesive strength of epoxy polymers depending on the heat treatment mode (Table 1)

The use of treatment mode No. 2 provides an increase in adhesive strength by 2-3% compared to the adhesive strength of epoxy polymers formed according to mode No. 1, since the duration of exposure at a temperature of 150°C increases to 5 hours. This makes it possible to ensure the formation of additional physical and chemical bonds between the components of the epoxy polymer matrix. Exposure within this regime for 2 hours at a temperature of 100°C allows to increase the adhesive strength to 18.9 MPa, which is due to the redistribution of reactive groups in the epoxy polymer binder.

A significant increase in the studied characteristic occurs under the condition of increasing the temperature at the final stage to 170°C. This allows for a higher degree of structuring of the epoxy system due to the activation of the chemical reaction rate. An increase in the duration of exposure at a temperature of 100°C provides an increase in adhesive strength to 20.8 MPa, which allows to activate the mobility of segments of macromolecules of the epoxy binder.

The highest values of adhesive strength (25.4 MPa) have epoxy polymers formed according to mode No. 4. Under such conditions, the formation of the maximum number of physical and chemical bonds occurs, since the duration of heat treatment is 5 hours at a temperature of 170°C. Increasing the exposure time to 2 hours at a heat treatment temperature of 100°C provides an increase in adhesive strength to 25.4 MPa, which is associated with a high exposure time in the thermal field.

The use of a heat treatment temperature of 190°C leads to a sharp decrease in the studied characteristic, since destruction processes occur after structuring at the initial stage of forming epoxy composite products. Destruction processes are associated with the partial destruction of physical and chemical bonds.

The hardness and impact strength of glass plates were studied on epoxy composite plates containing up to 6 layers of glass fabric with a density of 140 g/m². The lowest hardness values (76 MPa and 80 MPa) have glass plates (Fig. 2), which are formed according to the heat treatment mode No. 1.

The low hardness can be explained by insufficient exposure in the thermal field, which does not allow obtaining a large number of physical and chemical bonds. In the case of forming epoxy composites at a temperature of 170° C, the hardness increases to 96 MPa and 110 MPa, which is due to the completion

of the structuring process of the epoxy polymer matrix within 5 hours at the optimal temperature. The formation of epoxy composites at a temperature of 190° C leads to hardness values of 105 MPa and 108 MPa. The decrease in the hardness of epoxy composites at such a final temperature is associated with an increase in the fragility of the system due to the destruction processes.

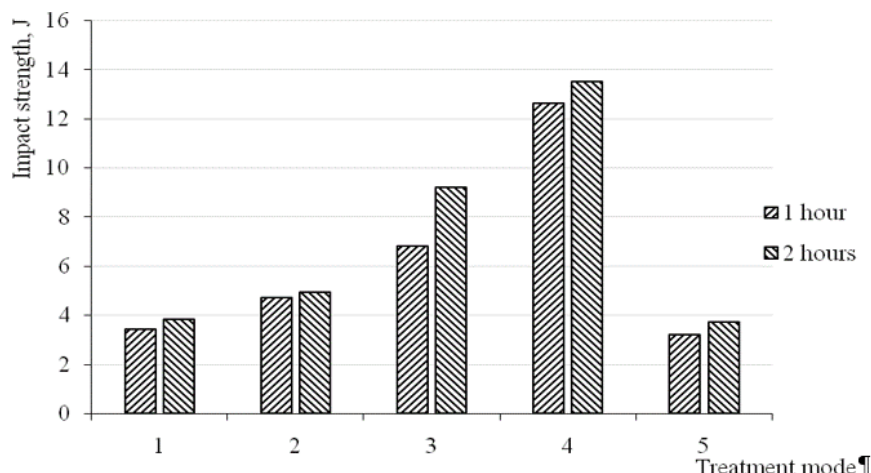


Fig. 2. Hardness of epoxy composites formed according to the different heat treatment regimes (Table 1)

The impact strength of epoxy composites formed by heat treatment mode No. 1 is 3.4 J and 3.8 J (Fig. 3) at different exposure times. This characteristic increases by 70-75% in the case using of heat treatment mode No. 4, which corresponds to exposure of epoxy composite samples at a temperature of 170° C for 5 hours. In this case, the epoxy polymer system is uniformly structured with the maximum number of physical and chemical bonds formed between the terminal active groups of the epoxy binder and the reactive groups –OH on the surface of the glass fibers.

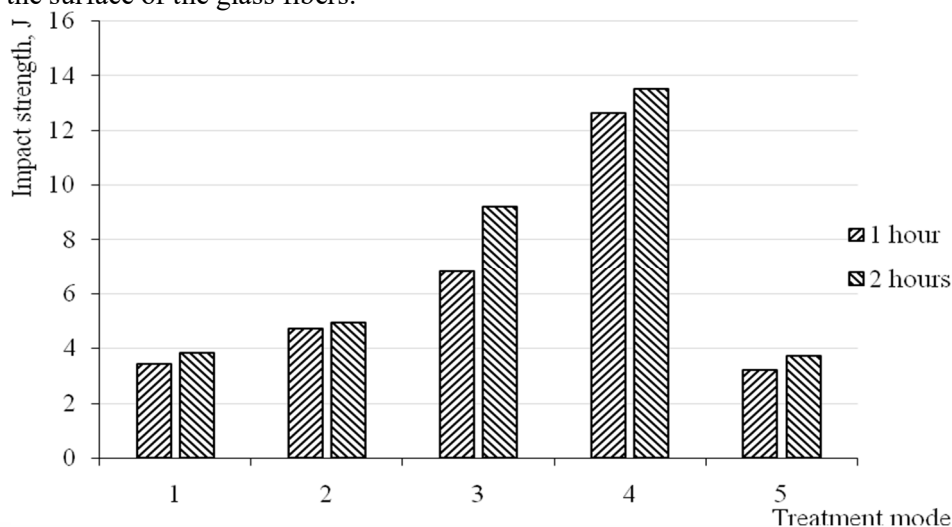


Fig. 3. Impact strength of epoxy composites formed according to the different heat treatment regime (Table 1)

The use of heat treatment at a temperature of 190° C leads to a sharp decrease in the impact strength of epoxy composites due to the destruction processes of the epoxy polymer matrix. The high structuring temperature leads to an uneven distribution of local volumes of the epoxy matrix, which are characterized by an increased degree of structuring of the system compared to the rest of the epoxy polymer volume.

The formation of a shear fracture zone (Fig. 4) occurs when a dynamic load is applied to an epoxy composite sample located on a steel disk.

There is a deformation zone without the destruction of the epoxy composite plate on the back side of the epoxy composite sample. This indicates a higher resistance of the material to the influence of dynamic loads (Fig. 5).

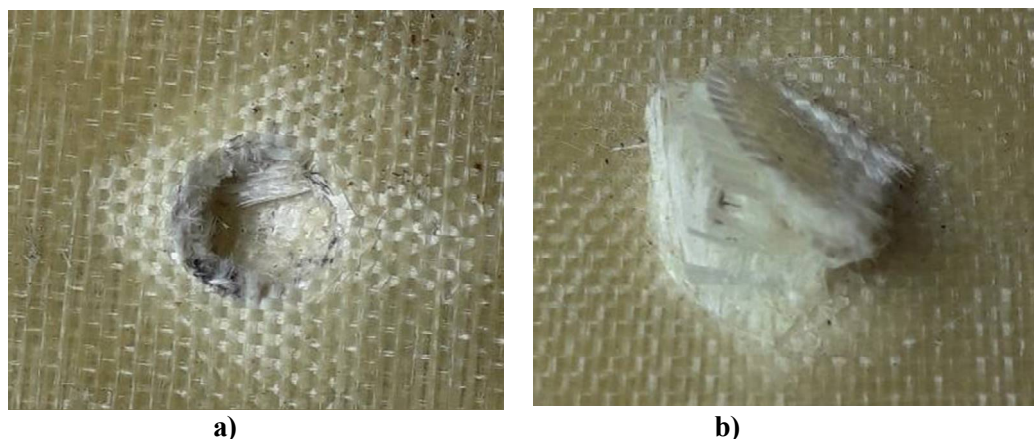


Fig. 4. General view of the fracture zone of the epoxy composite material formed according to the heat treatment mode No. 1 (1 h – 50 °C; 1 h – 100 °C; 3 h – 150 °C): a – front side of the sample; b – back side of the sample

The zone of delamination of the epoxy matrix from the glass fabric has a diameter of 17-18 mm, which indicates the propagation of a crack in the epoxy composite material. The delamination zone is smaller by 2-3 mm due to the increase in adhesion strength as a result of a longer exposure of 5 hour at a temperature of 150° C compared to the destruction of the epoxy composite plate, which is formed after exposure of 3 hour at a temperature of 150° C.

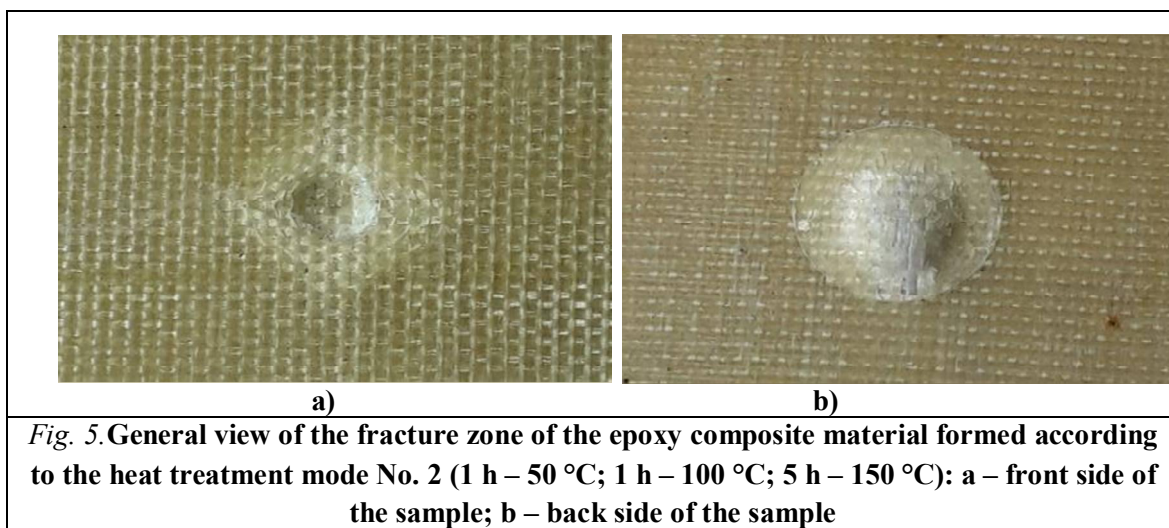


Fig. 5. General view of the fracture zone of the epoxy composite material formed according to the heat treatment mode No. 2 (1 h – 50 °C; 1 h – 100 °C; 5 h – 150 °C): a – front side of the sample; b – back side of the sample

When a dynamic load is applied to the epoxy composite plate, which is located on a steel disk, a depression is formed on the back side. The diameter of the delamination zone is 16-17 mm (Fig. 6). On the back side of the plate, the failure zone is a protrusion with through-hole damage to the epoxy composite material in a local zone with a segment of 50°. Such a material is capable of higher resistance to dynamic loads compared to epoxy composites treated according to modes No. 2 and No. 3.

On the front side, a peeling zone is formed without a significant degree of deformation of the sample on the steel disk (Fig. 7) in the case of applying a dynamic load to the epoxy composite plate. The diameter of the peeling zone on the front side is 11-12 mm. The diameter of the peeling zone on the back side of the plate is 15-16 mm. The main indicator of the resistance of the epoxy composite material to the influence of dynamic load is the absence of through-hole fracture of the epoxy composite plate, which indicates the effectiveness of heat treatment according to mode No. 4 (temperature 170° C, holding time 5 hours).

The destruction of the sample with the formation of a zone of through-fracture (Fig. 8, a) occurs in the case of applying a dynamic load to an epoxy composite plate containing aramid fabric (6 layers). This indicates a low resistance of aramid fibers to resist fracture under the influence of dynamic load compared to the impact strength of epoxy composites containing glass fabric with a density of 140 g/m².

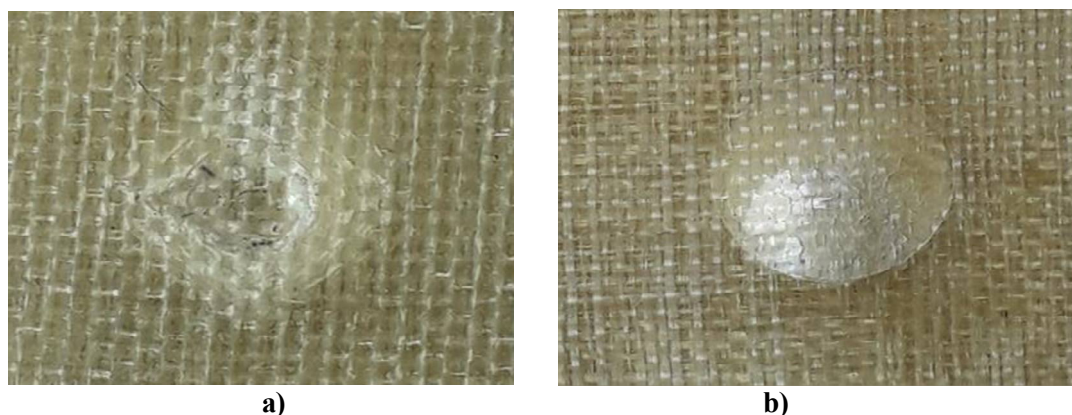


Fig. 6. General view of the fracture zone of the epoxy composite material formed according to the heat treatment mode No. 3 (1 h – 50 °C; 1 h – 100 °C; 3 h – 170 °C): a – front side of the sample; b – back side of the sample

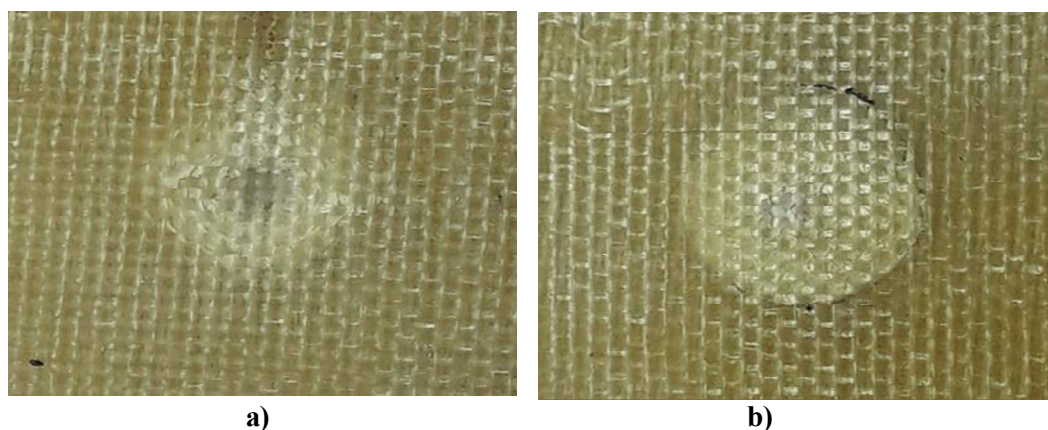


Fig. 7. General view of the fracture zone of the epoxy composite material formed according to the heat treatment mode No. 4 (1 h – 50 °C; 1 h – 100 °C; 5 h – 170 °C): a – front side of the sample; b – back side of the sample

The impact strength of epoxy composite materials containing 6 layers of fiberglass with a density of 600 g/m² is significantly higher (Fig. 8, b) compared to glass fabric based on epoxy polymer, which is filled with aramid fabric or glass fabric of lower density (140 g/m²). In this case, there is no zone of through-fracture, and there are no traces of plastic deformation in the zone of application of dynamic load.

Conclusions and prospects for further research. Epoxy polymers formed under the optimal heat treatment regime (170° C, exposure time 5 h) exhibit the highest adhesion strength of 25.4 MPa. Additional physicochemical bonds are formed between the active groups of epoxy resin macromolecules and hardener with an increased duration of heat treatment, due to the greater thermal energy absorbed by the epoxy composition compared to fiberglass materials formed in a thermal field over 3 hours. The hardness increases to 96 MPa and 110 MPa as a result of the formation of epoxy composites at a temperature of 170° C due to the completed structuring process of the epoxy polymer matrix after 5 hours at the optimal temperature.

Structuring the epoxy-polymer composition under the optimal heat treatment regime ensures the formation of an epoxy composite material with a high impact strength of 13.5-14 J. This treatment results in a uniformly structured epoxy polymer system, where the maximum number of physical and chemical bonds are formed. These bonds occur between the terminal active groups of the epoxy binder and the reactive -OH groups on the surface of the glass fibers.

Epoxy composite materials containing six layers of glass fabric provide resistance to dynamic loads, as they exhibit no zones of plastic. The developed epoxy fiberglass materials using glass fabric with a density of 600 g/m² and a number of layers of more than six is advisable to use for the manufacture of individual ballistic protection elements, as these materials resist penetration by foreign bodies with an impact energy of 24-26 J.

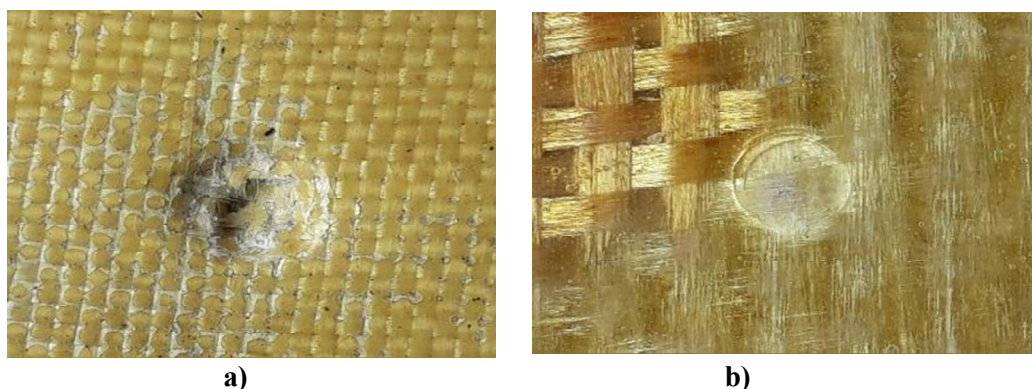


Fig. 8. Epoxy composites with filler content: a – aramid fabric; b – glass fabric (density 600 g/m²)

In the future, it is planned to conduct a study of the influence of modifiers on the formation of the epoxy polymer matrix and the impact toughness of epoxy composite materials.

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