INFLUENCE OF FILLER CONTENT ON MECHANICAL PROPERTIES OF ALUMINIUM AL99.5 SINGLE-LAP BONDS BONDED WITH ALUMINIUM AND POLYMER POWDER FILLED EPOXY ADHESIVE

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Abstract
An adhesive bonding technology is a promising method of a bonding. Aluminium ranks among significant technical materials, which are suitable for the adhesive bonding. The paper deals with the testing of composite materials based on aluminium and polymer microparticles. The aim of the research was to determine an influence of a content of a microparticle filler on mechanical properties of the polymer particle composite and adhesive bonds of the adhesive bonded material Al99.5. A tensile strength as well as a strength of the adhesive bond did not change when adding the filler. The highest increase of the tensile strength of 23 % (42.18 ± 1.92 MPa) was at the composite created from 2.5 g of the filler: 100 g of the matrix. The adhesive bond strength was increased when adding the filler except for the concentration 25 g of the filler: 100 g of the matrix (a decrease of 22 %). The adhesive bond strength increase was the highest at the adhesive bond with the adhesive in the form of the composite (2.5 g of the filler: 100 g of the matrix).The adhesive bond strength increase was 25.7 %. Adhesive bonds evinced an adhesive type of a fracture surface. By adding the filler in a form of microparticles there was not a change of the failure type. Using the electron microscopy within the experimental research the presence of cracks in the boundary adherent / adhesive (the matrix – two-component epoxy adhesive and the composite adhesive) at the adhesive bonds was proved.

Key words: adhesive bond strength, SEM, microparticles filler, tensile strength.

INTRODUCTION

A requirement for a mass decreasing of the construction is put when designing constructions of cars or agricultural machines etc. (BORSELLINO ET AL., 2009; MÜLLER, 2013). This requirement is secured namely by a use of aluminium materials in the constructions of machines. The aluminium constructions are lighter than traditional steel ones. However, the aluminium is of worse mechanical properties than the steel. The adhesive bonding technology is an effective method for the connecting of the aluminium and its alloys. The adhesive bonding technology advantage is a possibility to introduce an automation in the production process (SADEK, 1987). The use of the adhesive bonding technology in the construction of transport means and agricultural machines secures a stiffness comparable with mechanical fasteners or spot-welds. Further, the adhesive bonding technology increases an energy absorption reducing a noise and vibrations (BORSELLINO ET AL., 2009; MÜLLER, 2013).

However, the adhesive bonding technology has also its disadvantages, e.g. the strength of the bond depends on a choice of the adhesive, an overlapping geometry, a service life of the bond is limited etc. These properties can significantly influenced the final bond (BORSELLINO ET AL., 2009).

In the area of the bonding of sheets of metals, e.g. in the construction of agricultural machines, single-lap adhesive bonds are used. Technologies such as an adhesive bonding, a riveting and a welding are particularly used in manufacturing corporations focused on bonding of the metal sheets. These methods are frequently combined (MÜLLER, 2013).

Epoxies are widely used as high-performance structural adhesives. Epoxy resins are attractive for metal-bonding adhesive systems. Epoxies are able to bond well a variety of treated or untreated metal surfaces (RAMAZAN ET AL., 2008). Epoxy adhesives have a good affinity for aluminium alloy surfaces and the oxide layers produced during a surface preparation (CHASSER ET AL., 1993). A substantial change in mechanical properties can be achieved by adding an optimum volume of a filler i.e. a reinforcement. A composite material is created by this way. Composite materials are promising structural components (MÜLLER ET AL., 2015; RUGGIERO ET AL., 2015A; MÜLLER, 2011). They comprise one or more discrete

phases stored in the continuous phase (VALÁŠEK AND MÜLLER, 2015; VALÁŠEK ET AL., 2015). They can be used as the adhesive or a cement.

Polymers are important matrix materials for forming of advanced composites. The reinforcing materials in advanced composites take many forms. In addition to continuous fibres, many types of short reinforcing elements are used in composites. One popular form of short ‘‘fibres’’ is a spherical particle. A number of studies in particle composites have been undertaken with microparticles (CHO ET AL., 2006).

The filler of composite materials is different: Al_2O_3, SiC, glass beads, minerals, various metals, rubber particles. Mechanical properties of polymer composites strongly depend on the particle size (SHAO YUN FU ET AL., 2008; MÜLLER 2011). The type and the size of the filler depend on an application of the composite. A mild increase of the adhesive bond strength can occur by adding the filler (MÜLLER ET AL., 2015).

It was ascertained that an interfacial fracture toughness does not depend on the particle size but it increases substantially when the sliding fracture mode prevails (RAMAZAN ET AL., 2008; CHO ET AL.,. 2006).

The tensile strength of particle composites can be improved with decreasing particle size. However, composites with 3 vol% of nanoparticles resulted in lower tensile strength due to the likely poor dispersion of nanoparticles at higher particle loading than that ones with microparticles (CHO ET AL., 2006).

The finite element analysis results on stresses show that the aluminium content in the adhesive adversely affects the mechanical strength of the bond. However, a promising result was obtained through an experimental investigation that the epoxy adhesive retains its adhesion strength even with as much as 50 wt% addition of the aluminium filler. Even though the finite element analysis shows higher stresses at the adhesive–metal substrate interface, the actual failure occurs within the adhesive indicating that the strength of the adhesion to the metal substrate surface is stronger than the strength of the adhesive itself (RAMAZAN ET AL., 2008).

The aim of the research was to determine the influence of the content of the microparticle filler on the mechanical properties of the polymer particle composite and the adhesive bond of the bonded material Al99.5.

MATERIALS AND METHODS

In order to understand the size effect of all scales in particle composites, the failure mechanism and mechanical properties in these composites have to be understood. This study presents the laboratory experiment results performed on aluminium Al99.5 with the polymer composites reinforced with the aluminium microparticles (a multi-plate shape) and the plastics (a spherical shape). Mechanical properties of the composites were measured through tensile tests.

The objective of this study was to develop an influence of the aluminium and plastic filler content on the mechanical properties of the adhesive bonds bonded by the two-component epoxy ChS Epoxy 1200 (hardener P11 – Diethylentriamin).

The concentration of the filler in the matrix is indicated by the wt. fraction of the filler. The determination of the concentration of the sub-components was expressed using a weight relative to 100 g of the matrix (the two-component adhesive).

The filler was added into the matrix ChS Epoxy 1200 (two-component reactoplastics resin) in the ratio of 0.5, 1, 2.5, 10, 15, 20, 25 wt% to 100 wt% of the matrix. Weight percentages were chosen with a respect to a practical application when the filler is mixed mainly on the basis of weight ratios.

The adhesive bond strength was determined using the single-lap shear test according to CSN EN 1465. Laboratory experiments were performed on normalized testing samples of the aluminium Al99.5 prepared under the standard CSN EN 1465 by cutting the metallurgical semi-finished product in a form of the metal sheet.

The testing samples without a mechanical and a chemical treatment of the surface were used for the adhesive bonding. The untreated samples were used due to minimizing the factors effecting the preparation of the bonded surface. This trend is significant particularly in operations where the automation is implemented (NOVÁK, 2012; HRICOVA 2014; RUGGIERO ET AL., 2016; RUGGIERO ET AL., 2015B; VESELÁ ET AL., 2013; ALEŠ ET AL., 2012).

The roughness parameters Ra and Rz were measured on the adherent surface designated for the adhesive bonding. Roughness parameters were measured with the portable profilometer Mitutoyo Surftest 301. A boundary wave length of cut-off was placed to 0.8 mm.

The test specimens of the matrix and the composite materials for the tensile properties determination according to the standard CSN EN ISO 527-1 (Plastics – Determination of tensile properties – Part 1: General principles) were prepared according to the standard CSN EN ISO 3167 (Plastics – Multipurpose test specimens, Czech Standard Institution). By mixing of

the specified matrix – filler phases ratio the composite was made, which was used for the preparation of test specimens according to the specified standards. The moulds for casting were made from the material Lukapren N using models. The form and the size of moulds meet the corresponding standards.

The testing sample of the matrix and the composite materials was kept under the laboratory temperature 22 ± 2 °C for 48 hours after the fixation of the adhesive bond. A constant thickness of the adhesive layer was secured by the weight of 750 g. Afer that the destructive testing followed. The adhesive layer thickness was determined by the optical analysis of the adhesive bond cut.

The tensile strength and the adhesive bond strength were performed using the universal tensile strength testing machine LABTest 5.50ST (a sensing unit AST type KAF 50 kN, an evaluating software Test&Motion). A speed of the deformation corresponded to 10 $mm.min^{-1}$. The optical analysis of fracture surfaces and the adhesive bond cut was examined with SEM (scanning electron microscopy) using a microscope MIRA 3 TESCAN. The fracture surfaces were dusted with gold.

The effect of the microparticle dispersion on the composite failure was studied with SEM (scanning electron microscopy) images. The effect of the particle size, the wettability and the porosity on the interfacial crack was investigated with SEM.

An evaluation of the shape and the dimension was performed using the program GWIDDION. The results of measuring were statistically analysed. Statistical hypotheses were also tested at measured sets of data by means of the program STATISTICA. A validity of the zero hypothesis (H_0) shows that there is no statistically significant difference ($p > 0.05$) among tested sets of data. On the contrary, the hypothesis H_1 denies the zero hypothesis and it says that there is a statistically significant difference among tested sets of data or a dependence among variables ($p < 0.05$).

RESULTS AND DISCUSSION

The surface roughness of the tested specimens made by casting from the matrix was Ra 0.24 ± 0.06 μm, Rz 1.44 ± 0.47 μm. The surface roughness of the tested specimens made by casting from the matrix and the filler, i.e. the composite was Ra 0.23 ± 0.05 μm, Rz 1.40 ± 0.41 μm.

The matrix, i.e. the two-component structural adhesive showed the tensile strength 34.21 ± 3.01 MPa. It is obvious from the results of the strength of the matrix and the composite materials of various filler concentrations that adding the filler changes the tensile strength (Fig. 1). The highest increase of the tensile strength of 23 % (42.18 ± 1.92 MPa) was at the composite from 2.5 g of the filler: 100 g of the matrix.

It is possible to say in terms of the statistical testing of the influence of various filler concentrations on the tensile strength that they are statistically non-homogeneous groups. The hypothesis H_0 was not certified, i.e. there is a difference in the tensile strength in 0.05 significance level among single tested materials, i.e. the matrix and various concentrations of the filler ($p = 0.0000$).

The presence of diversely large filler particles was proved using the electron microscopy within the experimental research (Fig. 2). The aluminium microparticles were of the multi-plate shape. The plastics microparticles were of the spherical shape.

The considered particle sizes reached 14.71 ± 7.81 μm (aluminium length), 2.12 ± 1.03 μm (aluminium width) and 23.86 ± 18.62 μm (polymer – spherical particles).

The non-homogeneity of the adhesive is also evident from the fracture surface (Fig. 3). This is caused by air bubbles arisen during both the mixing process of the two-component epoxy adhesive and the hardening without using vacuum.

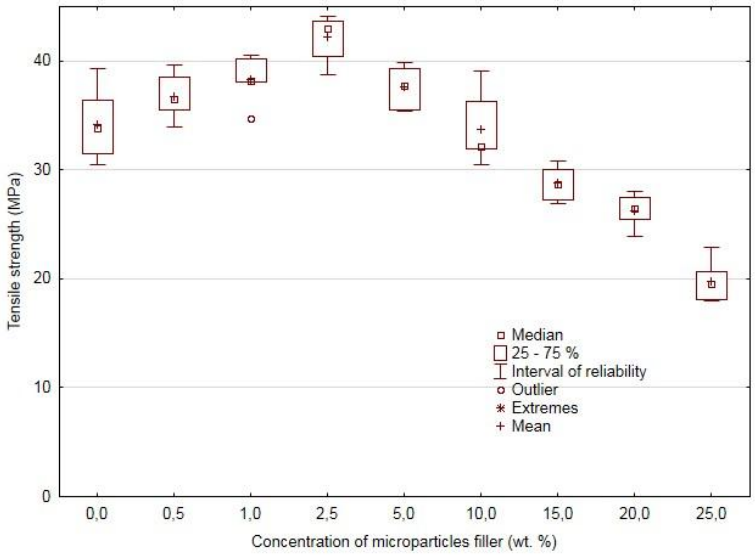

Fig. 1. – Influence of filler concentration on tensile strength

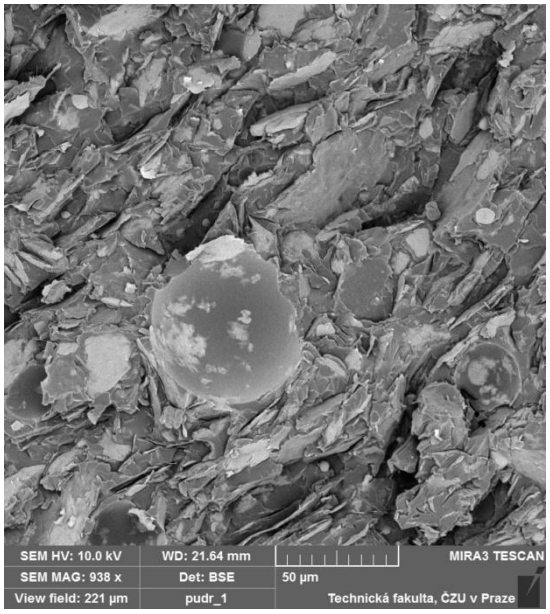

Fig. 2. – SEM images of composite material - microparticles of aluminium (multi-plate shape) and polymer particles (spherical shape)

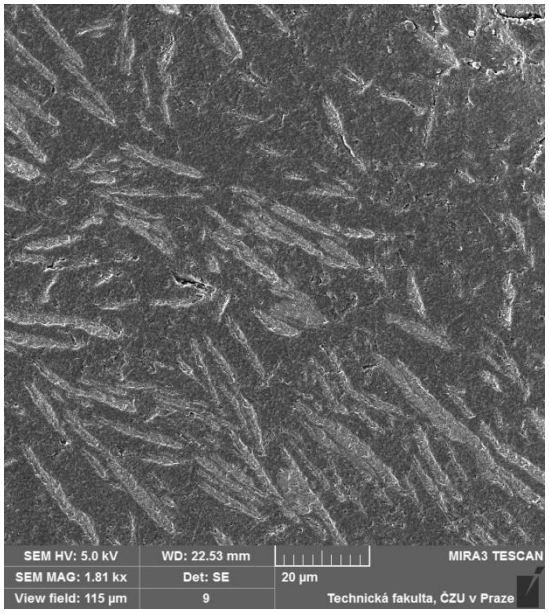

Fig. 3. – SEM images of cut through composite layer containing aluminium microparticles (multi-plate shape) and porosity

Fig. 4 and 5 show a histogram of a frequency of aluminium microparticles. It is obvious from the results that the highest portion was at the aluminium microparticles among 5 to 15 µm (a length) and 1 to 3 µm (a width).

The surface roughness of the adhesive bonded material Al99.5 was in the direction parallel to the loading force at the destructive testing of the adhesive bonds Ra 0.17 ± 0.02 µm, Rz 1.00 ± 0.28 µm and in the direction perpendicular to the loading force at the destructive testing of the adhesive bonds Ra 0.26 ± 0.02 µm, Rz 1.50 ± 0.20 µm.

The adhesive layer thickness was measured as 171.22 ± 21.97 µm. The optimum shear strength was reached at the two-component structural epoxy adhesives in the interval of the adhesive layer thickness 0.1 to 0.25 mm (MÜLLER AND VALÁŠEK, 2013).

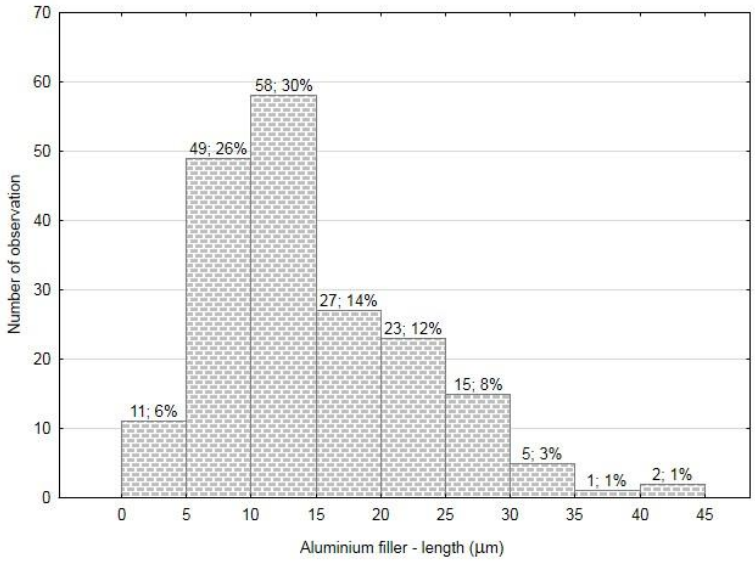

Fig. 4. – Histogram of length of filler in form of aluminium microparticles

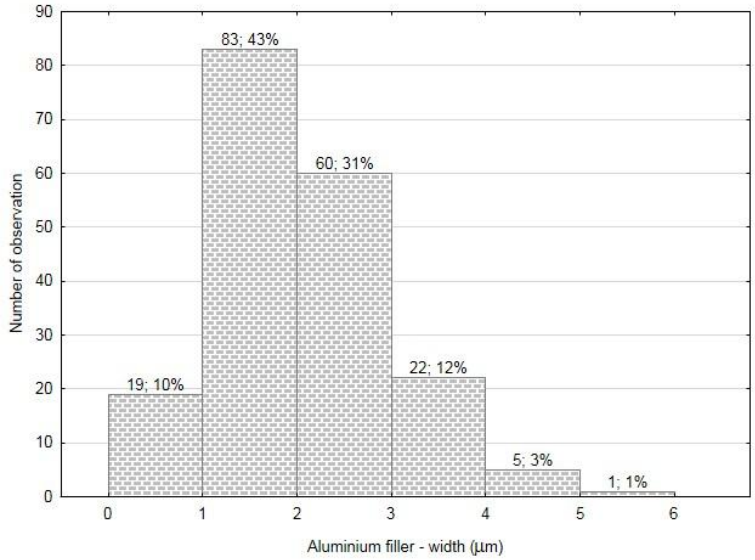

Fig. 5. – Histogram of width of filler in form of aluminium microparticles

It is obvious from the adhesive bond strength results that adding the filler changes the adhesive bond strength (Fig. 6). The highest adhesive bond strength increase was at the adhesive bond with the adhesive in the form of the composite (2.5 g of the filler: 100 g of the matrix). The adhesive bond strength increase was 25.7 %.

It is possible to say in terms of the statistical testing of the influence of various filler concentrations on the adhesive bond strength that concentrations are statistically non-homogeneous groups. The hypothesis H_0 was not certified, i.e. there is a difference in the adhesive bond strength in 0.05 significance level among single tested materials, i.e. the matrix and various concentrations of the filler ($p = 0.0001$).

Adhesive bonds evinced an adhesive type of the fracture surface. There was no difference between the adhesive bonds without the filler (the matrix) and with the filler.

It came to the increase of the adhesive bond strength but the adhesion of the adhesive was not increased owing to adding the aluminium microparticles.

A presence of cracks in the boundary adherent / adhesive was proved using the electron microscopy within the experimental research (Fig. 7). The adhesive failure of the adhesive bonds originated also in these cracks. It came to decreasing of the adhesive strength of the adhesive bond.

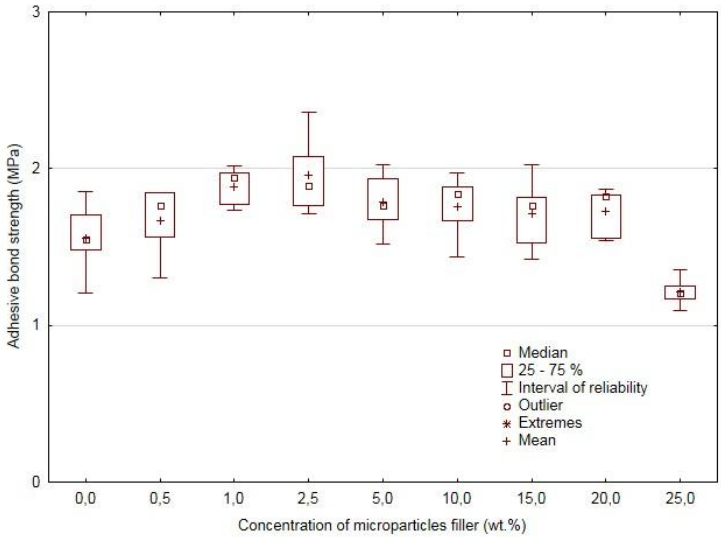

Fig. 6. – Influence of filler concentration on adhesive bond strength (adherent Al99)

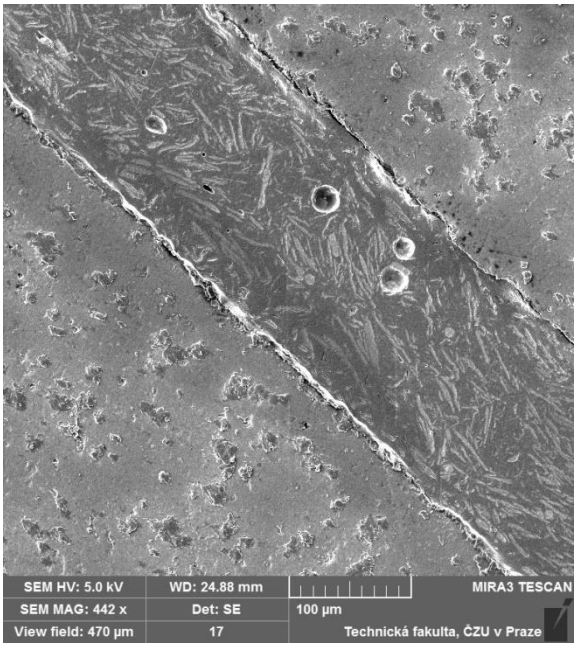

Fig. 7. – SEM images of microcracks in boundary adherent Al99.5 / adhesive (polymeric particle composite 1 g filler: 100 g matrix, without surface treatment of adherent)

The experiment results proved a bad wettability between the adhesive layer and the adherent. A weak interfacial bond was measured as 2.13 ± 1.56 µm. The research results proved a good wettability between the adhesive and the filler.

From the results of the experiment it is possible to agree with the statement that epoxy adhesives preserve their adhesive bond strength also at high concentrations of the filler (RAMAZAN ET AL., 2008).

A strong interaction between adhesive and particles is evident. When applying the filler into the resin, the wetting of the filler with the matrix is very important (JACKEL AND SCHEIBNER, 1991). Results of the experiment also show the irregular stratification of filler microparticles in the matrix (Fig. 3). CHANG ET AL. (2001) proved in their experiments that the irregular shape of the particles ensured good interaction between the matrix and the filler. The assumption about a negative influence of the filler on the tensile strength was not confirmed. CHO ET AL. (2006) state that there is a decrease in the strength of the composite with increasing volume of filler particles.

Results of the experiments confirmed conclusions of RAMAZAN ET AL. (2008), where they state that there was a slight increase of the adhesive bond strength of testing samples from the aluminium alloy with adding the aluminium microparticles. Microparticles of Al_2O_3, SiC affect the tensile strength in a negative way (CHO ET AL., 2006).

By adding the filler in the form of aluminium microparticles there was not a change in a failure type. Therefore it is not possible to agree with the statement of RAMAZAN ET AL. (2008) that filled epoxy resin improved the adhesion to the bonded surface, i.e. there was not a cohesive failure of the adhesive bond (RAMAZAN ET AL., 2008).

CONCLUSIONS

Following conclusions can be deduced from the results of the experiment focused on the research on the influence of the filler concentrations in the form of the aluminium microparticles and polymer on the me-

chanical properties of the polymeric particle composite and adhesive bonds of the material Al99.5:

- The tensile strength of the composite material changed when adding the filler. The highest increase of the tensile strength of 23 % (42.18 ± 1.92 MPa) was at the composite from 2.5 g of the filler: 100 g of the matrix. The tensile strength got worse compared to the matrix in the form of the two-component epoxy adhesive at the composite from 10 g of the filler: 100 g of the matrix when adding the filler.
- The adhesive bond strength changed when adding the filler. The adhesive bond strength increased (except for the concentration 25 g of the filler: 100 g of the matrix - a fall of 22 %) when adding the filler. The highest increase of the adhesive bond strength was at the adhesive bond with the adhesive in the form of the composite (2.5 g of the filler: 100 g of the matrix). The increase of the adhesive bond strength was 25.7 %.
- The adhesive bonds evinced an adhesive type of the fracture surface. The failure type did not change when adding the filler in the form of the microparticles.

The presence of cracks was proved at the adhesive bonds in the boundary adherent / adhesive (the matrix – the two-component epoxy adhesive and the composite adhesive) using the electron microscopy within the experimental research.

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