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COMPOSITES THEORY AND PRACTICE

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THE EFFECT OF ADHESIVE TYPE ON STRENGTH OF INTER-LAYER JOINTS IN FIBER METAL LAMINATE COMPOSITES

The paper presents the results of an experimental study concerning the strength of adhesive joints between the layers of a hybrid fiber metal laminate (FML) composite. The research was conducted on composites composed of aluminum 2024-T3 sheet metal and a glass fiber reinforced polymer (GFRP) prepreg made using the autoclave process. The key factor determining the quality of the layered composites is the high strength adhesive joint between the layers. The article discusses the issue of static inter-layer adhesive joint strength under different directions of loading for various types of adhesives. Shear strength tests for a single-lap joint were performed, as well as the peel strength test using the drum peel test. The strength tests were conducted for the variant that had an inter-layer joint made by epoxy included in the prepreg, while the second variant used an additional layer of adhesive film. The surfaces of the metal layers were prepared in accordance with the methodology used in aerospace production processes. The sheet surfaces were anodized in a sulfuric acid solution and then primed. Surface structure measurements of the sheets were made immediately before the joining process. Each layer was assembled in a clean room. The strength tests of the adhesive joints were conducted in static shearing and peeling conditions at room temperature. The results show that under shear loading the adhesive film lowers the elastic module of the joint and results in a slight increase in strength. However, under normal loading, there was 289.4% increase in the peel strength of the joint with the adhesive film. After the strength tests the surfaces of the destroyed adherends were analyzed using SEM. For the shear strength specimens no significant differences were found, whereas for the specimens subjected to peeling it was shown that cohesive damage was observed for the variant with the adhesive film, while the specimens without adhesive film were characterized by adhesive damage.

Keywords: **FML composites, adhesive joints, shear strength, peel strength, 2024-T3 aluminum alloy, GFRP, adhesive film**

WPŁYW RODZAJU SPOIWA NA WYTRZYMAŁOŚĆ MIĘDZYWARSTWOWEGO POŁĄCZENIA ADHEZYJNEGO W KOMPOZYTACH FML

Przedstawiono wyniki badań eksperymentalnych dotyczących wytrzymałości połączenia adhezyjnego pomiędzy warstwami składowymi hybrydowego kompozytu metalowo-włóknistego (FML). Próby przeprowadzono dla kompozytów będących połączeniem blachy ze stopu aluminium 2024-T3 oraz kompozytu polimerowo-włóknistego (szklanego) wykonanych w procesie autoklawowym. Kluczowym czynnikiem determinującym jakość kompozytów warstwowych jest wysokiej wytrzymałości połączenie adhezyjne pomiędzy warstwami. Artykuł porusza zagadnienie wytrzymałości statycznej połączenia międzywarstwowego przy różnym kierunku obciążenia dla różnego typu spoiwa. Przeprowadzono badania wytrzymałości na ścinanie na podstawie wytrzymałości połączenia jednozakładkowego oraz badania wytrzymałości na oddzieranie na podstawie próby tzw. „drum peel test". Badania wytrzymałości przeprowadzono dla wariantu, w którym połączenie międzywarstwowe uzyskano poprzez żywicę epoksydową będącą syciwem prepregu, natomiast w drugim wariancie zastosowano dodatkową warstwę kleju błonkowego. Powierzchnie warstw metalowych przygotowano zgodnie z metodologią stosowaną w przemyśle lotniczym. Powierzchnie blach anodowano w roztworze kwasu siarkowego, a następnie gruntowano. Przeprowadzono trójwymiarową analizę struktury powierzchni bezpośrednio przed procesem łączenia poszczególnych warstw kompozytu. Warstwy kompozytu układano we właściwej konfiguracji z zachowaniem zasad czystości w pomieszczeniu „czystym" (tzw. clean room). Przeprowadzono badania wytrzymałości statycznej połączenia adhezyjnego na ścinanie oraz oddzieranie w temperaturze pokojowej. Badania wykazały, że przy obciążeniach stycznych klej błonkowy wpływa na obniżenie modułu sprężystości połączenia oraz nieznacznie na wzrost wytrzymałości. Natomiast przy obciążeniach normalnych wykazano wzrost o 289,4% wytrzymałości na oddzieranie połączenia w wariancie z klejem błonkowym. Po przeprowadzonych badaniach wytrzymałościowych powierzchnie klejone poddano analizie przy użyciu m.in. mikroskopii skaningowej SEM. Dla próbek poddanych badaniom wytrzymałości na ścinanie nie wykazano tu istotnych różnic, podczas gdy próbki poddane oddzieraniu cechowały się istotnymi różnicami zależnie od wariantu. Dla próbek z klejem błonkowym wykazano zniszczenie kohezyjne, natomiast dla próbek bez kleju błonkowego - zniszczenie adhezyjne.

Słowa kluczowe: **kompozyty FML, połączenia adhezyjne, wytrzymałość na ścinanie, wytrzymałość na oddzieranie, stop aluminium 2024-T3, kompozyty polimerowo-włókniste, klej błonkowy**

INTRODUCTION

Composite materials are currently being used more widely, especially in aviation due to their obvious multiple advantages. However, they have several limitations that result in them not being used throughout the entire structure of the aircraft. The limitations primarily appear in places where the aircraft is exposed to high temperatures. As a result, there is a necessity to join polymer fiber composites with other materials, primarily metals like aluminum alloys and titanium [1]. Adhesive joints have an advantage over the process of riveting, which is popular in aircraft construction, because they do not require the drilling of holes that are frequently points of stress concentration and do not contribute as much mass to the total vehicle weight like rivets [1].

Most available literature on the issue of adhesive joint strength between aluminum alloy sheet metal and fiber reinforced polymer composites focuses only on the shear strength or cracking mechanisms. A double cantilever beam is most commonly used as the test subject for cracking analysis. The aim of this test is to determine the behavior of crack propagation and collect the energy data for numerical analysis of the cracking mechanisms [2, 3]. In turn, single or double-lap joints are most commonly used to determine the shear strength of the analyzed metal-composite joints [4-7]. Many authors have approached the analytical issue as well as numerical analysis related to the shear strength of adhesive joints between various materials [8-10]. The common use of adhesive joints in the aviation industry makes the fatigue strength of adhesive joints also a very important issue [11, 12].

When considering the issue of using adhesive to join material with significantly different coefficients of thermal expansion, the stress in the plane of the joint of these materials that results from various operating temperatures should be considered, which can contribute to reducing joint strength. Hence, many works focus on the possibility of minimizing the negative effects resulting from various material properties that are joined with a properly selected adhesive layer [13, 14].

Issues concerning adhesive joints between fiber composites and aluminum alloy sheet metal appear in the construction of FML composites and it can be stated that proper adhesive joining in the process of manufacturing the composite determines the further operational quality of the layered composite structure. FML composites made from prepregs based on epoxy resins are manufactured in such way that the adhesive joint between the layers is created by the resin that is the saturant of the prepreg [15]. Research has been conducted on this type of joint formation, while the results were compared with techniques that make adhesive joints using adhesive film as an additional binder between the prepreg and sheet metal. Adhesive films are used in composites that have a fiber layer based on a thermoplastic matrix [16].

The article presents the results from an experimental study determining the strength of adhesive joints between fiber reinforced polymer composites and 2024-T3 aluminum sheet metal. The most commonly analyzed forms of loading (shearing) adhesive joints were the focus of the study. Tests focusing on an undesirable form of loading known as peeling that is characterized by uneven stress distribution in the adhesive joint and stress accumulation on the face where the load is applied were also conducted.

EXPERIMENTAL PROCEDURE

Materials and methods

The layered composite used in the study was made of 2024-T3 aluminum alloy sheet metal. The adherend surfaces were anodized according to the following procedure. Oxide coatings were produced on the 2024-T3 substrate by means of the anodizing process. The specimens were abraded with grade 320 sand paper, rinsed with water and degreased in an NaOH aqueous solution (100 g·dm^{-3}) for 1 minute at 25°C, rinsed with deionized water, and etched in an HNO_3 aqueous solution (400 g·dm^{-3}) for 1 minute at 25°C. Subsequently, they were anodized in an H_2SO_4 aqueous solution (300 g·dm^{-3}) at 15°C. A constant current density equal to 1 A·dm^{-2} was applied. When the anodizing process was completed, the obtained coatings were rinsed with deionized water and dried in air. Their thickness was determined using the eddy-current method (Dualscope FMP100, Fischer). For a substrate thickness equal to 2 mm, the thickness of the coating was 10 ±1 μm, for a thinner substrate (0.4 mm) thinner coatings were produced (6 μm). The surfaces of the sheets were measured by a white light interferometer Talysurf CCI Lite using the 5x objective. The parameters of the surface textures were calculated using TalyMap software. The measured areas of 3.3 mm x 3.3 mm contained 1024 x 1024 points. The textures of the surfaces were only leveled, digital filtration was not used.

Figure 1 presents a three-dimensional structure of a fragment of the sheet metal that was adhered to the composite. Figure 1a presents the structure of 2 mm thick sheet metal, while Figure 1b presents the structure of the sheet metal with a thickness of 0.4 mm. Table 1 contains a list of roughness parameters of the surface of the sheet metal used for three-dimensional analysis.

After the anodizing process, the aluminum surfaces were primed with EC3924B (3M, Maplewood, Minnesota, USA) in a time frame no longer than 24 hours.

The base material for creating the fiber layer of the composite was the glass/epoxy woven HEXPLY-916G prepreg manufactured by Hexcel (Hexcel Corporation, Stamford, Connecticut, USA).

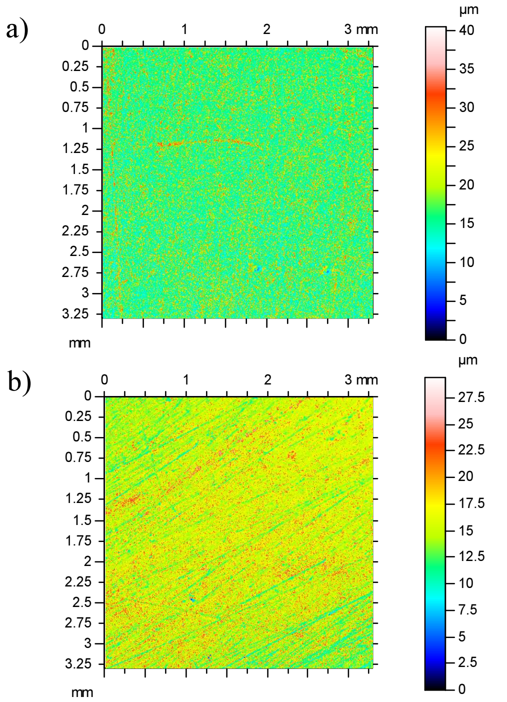

Fig. 1. Three-dimensional sheet metal surface structure before adhesion for 2 mm (a) and 0.4 mm (b) thick sheet metal

Rys. 1. Trójwymiarowa struktura powierzchni blach przed procesem klejenia: dla blachy o grubości 2 mm (a) oraz 0,4 mm (b)

TABLE 1. **Roughness parameters of sheet metal adhesion surface for three-dimensional analysis**

TABELA 1. **Parametry chropowatości powierzchni klejonych blach dla analizy trójwymiarowej**

Roughness parameters	0.4 mm thick sheet	2 mm thick sheet
S_q [µm]	2.72	4.32
S_{sk} [µm]	1.18	1.39
S_{ku}	4.53	3.75
S_p [µm]	13.7	22.9
S_v [µm]	15.8	17.6
S_z [µm]	29.4	40.5
S_a [µm]	1.97	3.36
S_{al} [mm]	0.0089	0.00726
S_{tr}	0.315	0.756
S_{td} [°]	34.8	84.5

The article presents a comparative study of two variants of an FML layered composite that differ in the method of creating the adhesive joint between the metal layer and the fiber reinforced polymer. The first variant is based on the common approach of manufacturing FML composites, which has a inter-layer joint made from epoxy resin that is the matrix of the prepreg (Fig. 2a). The second of the variants used an adhesive film as a intermediate element between the prepreg and sheet metal in order to create a high-strength adhesive joint between the layers (Fig. 2b).

In the variant with the adhesive film, Structural Adhesive Film AF-163-2K, which is a non-woven supporting carrier produced by 3M Scotch-Weld™, was used.

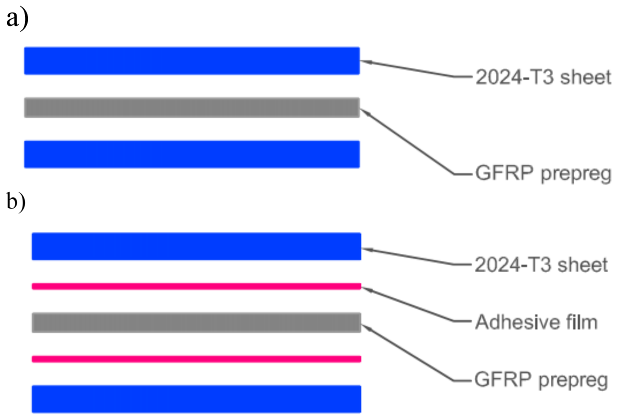

Fig. 2. Configurations of FML composite (2/1 lay-up) variants considered: a) without adhesive film, b) with adhesive film

Rys. 2. Konfiguracja kompozytu FML w wariancie 2/1 w rozważanych wariantach: a) bez kleju błonkowego, b) z klejem błonkowym

Sheets made with the appropriate layers were hardened in an autoclave using the following parameters: heating rate 2°C/min, curing temperature 135°C, pressure in the autoclave chamber during hardening 3 bar, pressure inside the vacuum bag –0.7 bar until reaching the crosslinking temperature, hardening time 90 min, cooling rate after hardening 3°C/min.

The composites intended for the study were manufactured as sheets of the following dimensions: shear test specimens - 200x360 (Fig. 3a) and peel test specimens - 300x360 (Fig. 3b).

From the aforementioned sheets, specimens were cut out to conduct the selected strength tests. In the case of the sheets intended for shear strength tests, the sheets were milled along the entire length on both sides in order to achieve an overlap. The specimens were cut out using a high pressure fricative water-jet generated indirectly. Machining of the specimens was performed at a pressure of p = 350 MPa, a grain flow rate of 300 g/min and nozzle feed rate of vf = 250 mm/min.

As a result, specimens for the selected strength tests were obtained. The static shear test of an adhesive joint between the sheet metal and glass/epoxy laminate was developed on the basis of standards regarding shear strength tests of overlapping adhesive joints of metal. The sheet was cut to make specimens with a width of 25 mm (Fig. 4a). The strength test was conducted on a Zwick/Roell Z100 strength test machine, a jaw feed rate of 2 mm/min was used.

For the peeling tests, a climbing drum peel strength test according to DTD 5577 was used. These tests were conducted on a universal Tinius Olsen H25K-T testing machine. 25 mm wide specimens with an adhesive joint length between the fiber reinforced composite and metal layers equal to 200 mm were made from a stiff

2 mm thick sheet (rigid adherent), while the flexible adherent was made of an 0.4 mm thick sheet (Fig. 4b).

Five specimens were tested and the average values of the mechanical parameters were determined.

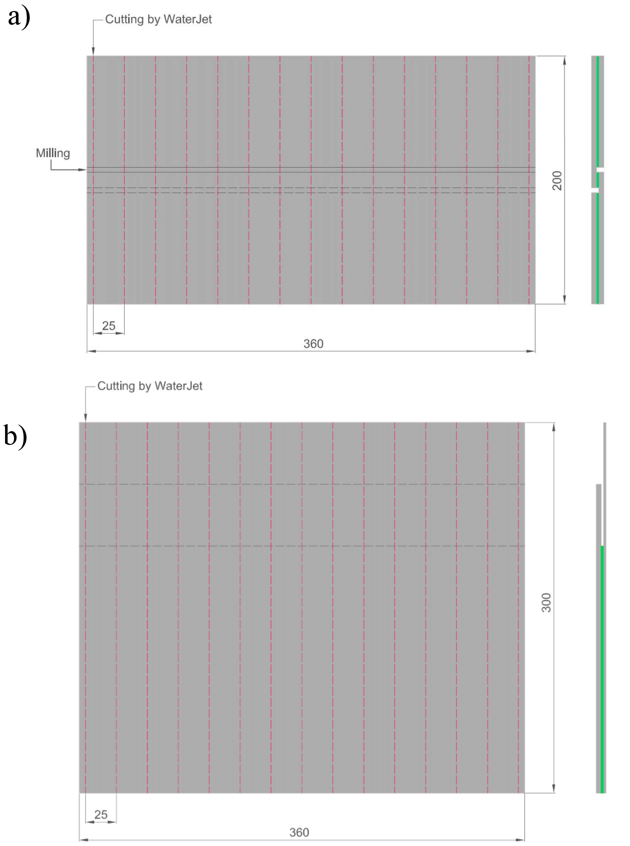

Fig. 3. Dimensions of sheet specimens with cutting direction for shear tests (a) and peel tests (b)

Rys. 3. Wymiary arkuszy próbek z przedstawionym kierunkiem rozcinania dla próby ścinania (a) oraz oddzierania (b)

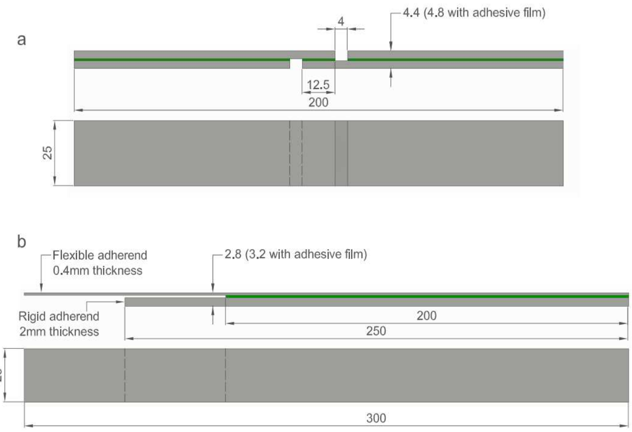

Fig. 4. Geometry of FML test specimens for: a) shear strength test and b) peel strength test

Rys. 4. Wymiary próbek do badań: a) wytrzymałości na ścinanie oraz b) oddzieranie

The morphology of the specimens after the tests as well as the chemical composition of their surfaces were determined by a scanning electron microscope (SEM) HITACHI S-3400N with an EDS/WDS system.

RESULTS AND DISCUSSION

Individual specimen variants underwent profile analysis to make the structural differences visually apparent for the versions with and without adhesive film. The images of the specimen profiles are presented in Figure 5. In the case of the variant with the adhesive film, the thickness of the fiber reinforced polymer composite layer is visibly larger.

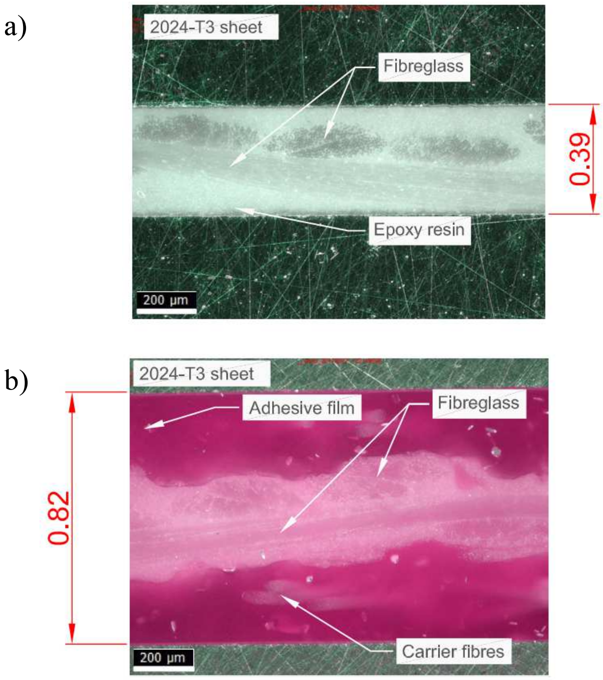

Fig. 5. Microstructures of studied composites: a) without adhesive film, b) with adhesive film

Rys. 5. Mikrostruktury stosowanych próbek kompozytowych: a) bez kleju błonkowego, b) z klejem błonkowym

Shear strength test

The shear strength tests revealed a comparable strength value for both the analyzed variants, which was on average 6.74 MPa for the specimens with the adhesive film and 6.52 MPa for the specimens made using only epoxy resin contained in the prepreg as a interlayer adhesive. The tests resulted in a high level of repeatability throughout the 6 repetitions. Figure 6 presents a tension curve of the specimens. When comparing both of them, it can be stated that those manufactured using adhesive film as an additional joint between the layers causes a slight increase in the elasticity of the joint.

Analyzing the surfaces of the adhesive joint after failure on the macroscale yields an adhesive failure mode. When analyzing the individual surfaces on the microscale, the images made using SEM reveal visible differences between the variants. In the case of the variant where adhesive film was not used (Fig. 7a), there are visible remnants of epoxy resin on the end parts of the adhesive surface on the uncovered sheet metal.

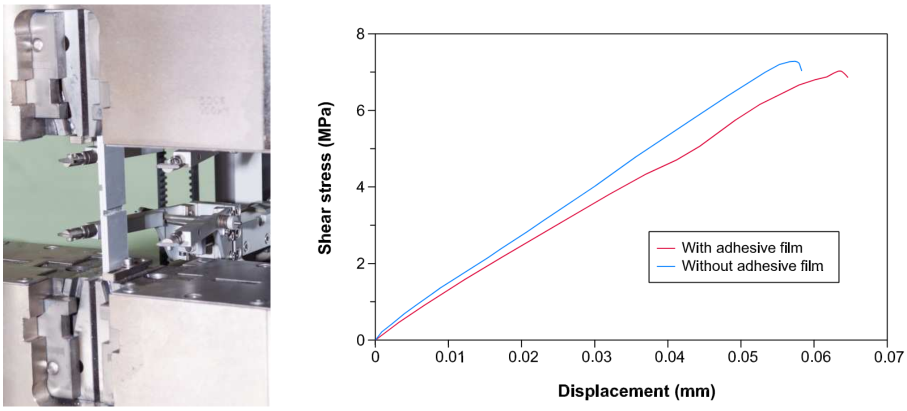

Fig. 6. Typical tensile/shear test curves of studied FML composites

Rys. 6. Typowe krzywe rozciągania uzyskane w badaniach wytrzymałości na ścinanie rozważanych kompozytów FML

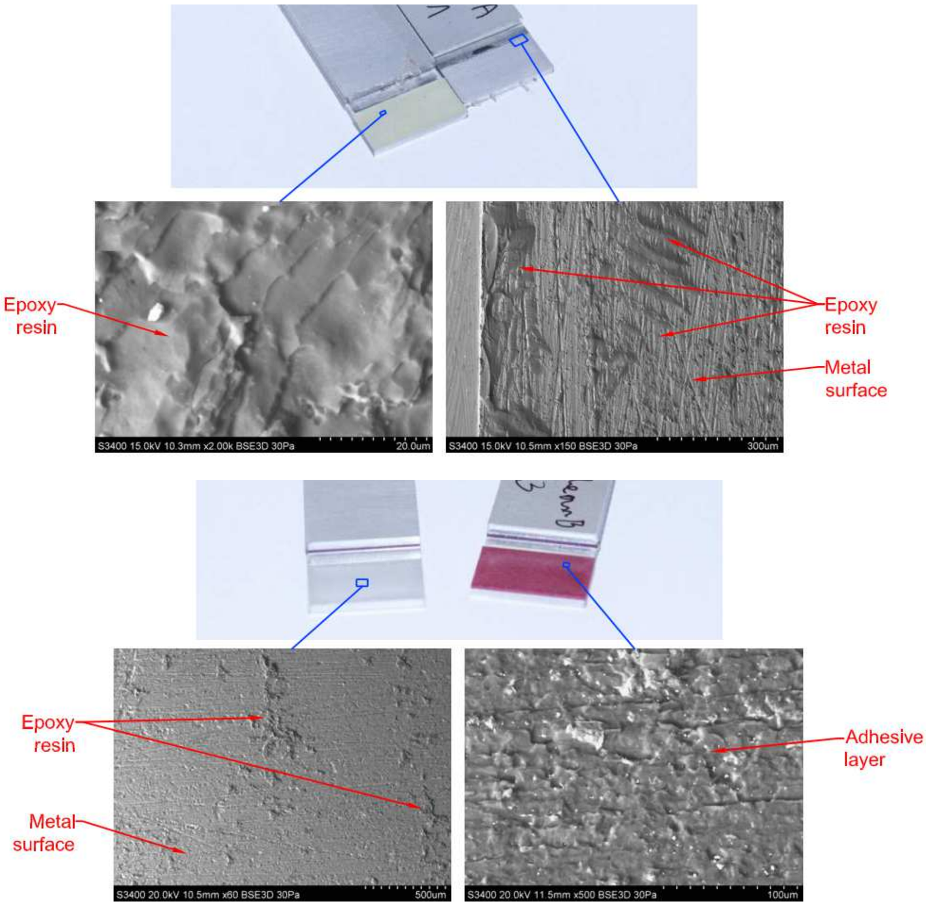

Fig. 7. SEM images of fractured surface after shear test: a) specimen without adhesive film, b) variant with adhesive film

Rys. 7. Obrazy wykonane mikroskopem skaningowym powierzchni po przeprowadzonych próbach ścinania: a) próbka bez kleju błonkowego, b) z klejem błonkowym

The remaining part of the sheet metal does not have any signs of resin. This may result from the reaction in the initial phase of the destructive process caused by the normal component of stress near the edge of the joint that may lead to tearing between the joined elements. However, the remaining part underwent traditional shear failure in the layer that joined the resin with the sheet metal. Thus, it can be said it was an adhesive failure mode.

In the case of the specimen with the adhesive film layer (Fig. 7b), it was noticed by means of microscopic observation that remnants of the adhesive covered the whole adhesion surface in a regular pattern where no fiber reinforced composite remained.

Peel strength test

The results of the peel test show that there are significant differences between the studied variants of joining composite layers. Figure 8 demonstrates example curves from the climbing drum peel strength test. Under stress loading conditions that are normal for an adhesive joint layer, the application of adhesive film caused a significant increase in strength. The average peel strength of the variant without adhesive film was 2.83 N/mm, while for the variant that included adhesive film the average result was 11.02 N/mm. The obtained increase in peel strength in the variant with adhesive film is a very desirable strength property because adhesive joints are typically poorly resistant to normal stresses.

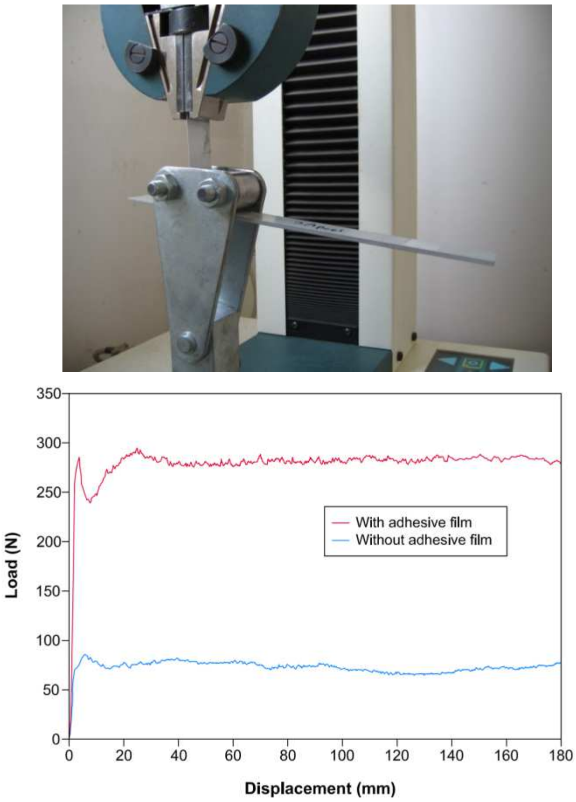

Fig. 8. Examples of load-displacement graphs measured during peel tests

Rys. 8. Typowe krzywe siła-przemieszczenie uzyskane w badaniach wytrzymałości na oddzieranie rozważanych kompozytów FML

When analyzing the surfaces of the specimens on the macroscale after conducting the strength tests, it can be noticed that there are significant differences in the failure modes. The specimens without adhesive film (Fig. 9a) demonstrated an adhesive failure mode for every test. Meanwhile, the adhesive film specimens (Fig. 9b) were characterized by cohesion failure on the entire separation surface, which confirms the high joint quality. When analyzing the SEM images, it can be noticed that minor remnants of epoxy resin are on some fragments of the surface of the specimens without adhesive film (Fig. 9a). However, the adhesive film specimens (Fig. 9b) have a noticeable microscopic surface resulting from adhesive failure modes. The separation surface displays uncovered fragments of the carrier fiber.

CONCLUSIONS

The results of the study describe the influence as positive as well as negative effects of applying an additional joining element to the adhesive joints of individual layers of FML composites. The primary advantage of using adhesive film is the significant increase in peel strength, which improved by 289.4%. This is an important property for this type of composite, even if adhesive joints should primarily withstand tangential stresses that cause shearing. However, in practice these joints are frequently loaded with normal stresses that cause separation or even peeling. Thus, the inclusion of adhesive film as an additional joining element can have a decisive effect on the layer strength of the structure.

A definite advantage of using an additional joining element can be a slight increase in joint elasticity, which in theory could be beneficial under cyclical variable loading. This more elastic joint form can lead to strength improvement and an increase in the fatigue life of the joint.

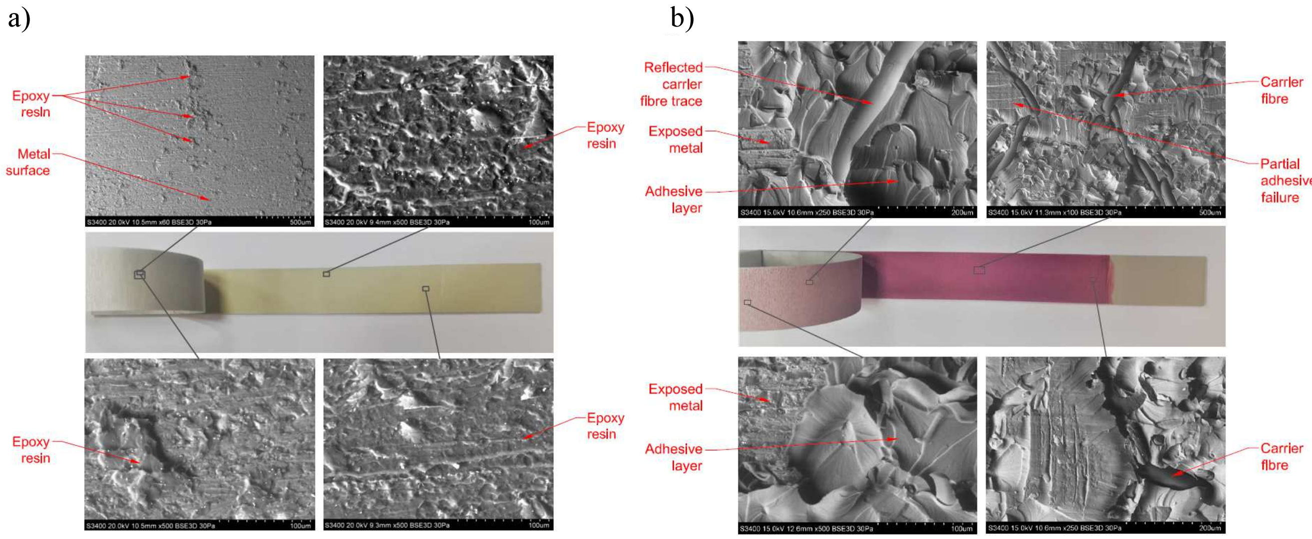

Fig. 9. SEM images of fractured surface after peel tests of specimen without (a) and with (b) adhesive film

Rys. 9. Obrazy wykonane mikroskopem skaningowym powierzchni po przeprowadzonych próbach oddzierania: a) próbka bez kleju błonkowego, b) z klejem błonkowym

When considering the disadvantages of additional joining elements, it should be noted that the presented composite configuration - 2/1 where there are two layers of adhesive film, increased the mass of the composite by about 10%. The addition also increases the thickness of the composite by about 30%. The last factor that needs to be mentioned is the economic one, including a double layer of adhesive film for one layer of prepreg increases the material costs by approximately 20%.

The results of the study represent one possible method of determining the static strength of adhesive joints that are increasingly common in advanced structures that require a compromise between light weight and high strength. Theoretically, applying this methodology to prepare the specimens should ensure high joint strength due to the application of the autoclave process. A logical continuation of the study will be a comparative study between the use of techniques with and without an autoclave. These trials are desirable because autoclave processes are both energy and work intensive, which results in higher costs. Developing alternative technologies that can ensure comparable joint strength are inevitable and the presented method could serve as a verification method of joint quality resulting from other techniques. In turn, the results from the study can serve as a reference point in comparative studies.

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