Study of the Behavior and the Mechanical Properties of Adhesively Bonded Polymer Matrix Composites Under Mechanical Loading

Daniela Spasova [1], Yaroslav Argirov [1], Plamen Petrov [1], Tatyana Mechkarova [1]

[1] *Technical University of Varna, "Studentska" Str. 1, Varna, Bulgaria*

Abstract **– Glass fiber reinforced Polymer matrix composites (PMCs) have a complex set of properties - high specific strength and hardness, good corrosion and chemical resistance, and most of all low relative weight, making them the preferred material over metallic materials in a number of industries (aircraft, boat building, renewable energy facilities and many others). The main aim set in the present work was reduced to the obtaining of PMCs made by adhesive bonding of two composites with a matrix of different types of resin (vinyl ester and epoxy) and the study of their behaviour in determining their mechanical properties. The studies were carried out with four types of adhesively bonded PMCs made in the form of laminates produced from a combination of three types of resin (two types of vinyl ester and one type of epoxy resin) and reinforced with biaxial fiberglass. The aim is to combine the lack of shrinkage of the epoxy resin with the better mechanical properties and better productivity of the vinyl ester resin. An analysis to determine shear strength, tensile strength and bending strength of the investigated composites was made. A macrostructural fractographic analysis to investigate the material behaviour under mechanical loads was carried out.**

Keywords **– Polymer Matrix Composites, epoxy resin, Vinyl ester resin, fiberglass, adhesive bonding**

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Corresponding author: Daniela Spasova,
Technical University of Varna, "Studentska" Str. 1, Varna, Bulgaria.
Email: danielats@tu-varna.bg

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1. Introduction

Polymer matrix composites (PMCs) have proven and established themselves in many industrial fields over the past decades. With the continuous development of composite materials, they are able to establish themselves in industries where their advantages are largely unknown. Glass fiber reinforced PMCs have a complex set of properties - high specific strength and hardness (similar to metallic materials), good corrosion and chemical resistance, and most of all low relative weight, making them the preferred material over metallic materials in a number of industries (aircraft, boat building, renewable energy facilities and many others) [1], [2], [3].

The design features of some PMCs require that they be made of separate elements that are subsequently joined to maintain their structural integrity in service under different mechanical loads. Typically, the joining of composite materials can be of two types- mechanical or adhesive bonding [1]. In a number of cases, adhesive bonding (which is based on a continuous bond based on cohesion, distributing the load over a larger area) is preferable to mechanical fastening, as drilling holes for bolts or rivets compromises the integrity of the reinforcing fibers and stress concentrators appear in the fastener hole [2], [4], [5].

There are a wide variety of different types of resins with characteristic properties distinguishing them from each other used as matrix material in the fabrication of PMCs. The most commonly used resins are: vinyl ester resin, epoxy resin and polyester resin. Vinyl ester resin and epoxy resin have better mechanical and performances than polyester resin, which is why the study conducted in this paper was limited to them. Fabrication of PMCs can be done in two ways- by vacuum forming or by hand-contact forming. It is characteristic of vinyl ester resin that it has better mechanical characteristics than epoxy resin but after curing 5÷6% shrinkage is obtained. Epoxy resin practically does not shrink after curing and it is not recommended for manual contact forming but mainly for vacuum forming, where surfaces with complex relief cannot be obtained.

Guided by the above features, the task set in the present work was reduced to the obtaining of PMCs made by adhesive bonding two composites with a matrix of different types of resin (vinyl ester and epoxy), in order to better functionality and performance, and the study of their behavior in determining their mechanical properties [6], [7]. Most commonly, failure analysis of adhesively bonded products is performed by interfacial failure under different mechanical loading conditions, based on the resulting strength of the products, which is also a criterion for the bonded surface strength. The failure and delamination of adhesively bonded joints (laminated composites) is due to both linear and non-linear deformations of the bonded elements [8], [9], [10]. The development of the crack can be reduced to a combination of three independent factors: I- crack opening - tensile stress normal to the crack plane; II – sliding mode - shear stress acting parallel to the crack plane and perpendicular to the crack front; III - rupture mode (shear stress acting parallel to the crack plane and parallel to the crack front) [11].

In order to analyze the adhesively bonded surface strength and the behavior of the adhesively bonded PMCs under the influence of mechanical loading and taking into account the specificity of the crack formation and propagation mechanism, mechanical tests determining their shear strength, tensile strength and bending strength were conducted.

2. Experimental Procedure

The studies were carried out with four types of adhesively bonded PMCs (Table 1) made by hand-contact method in the form of laminates, by layer-by-layer application of resin (matrix) and fiberglass fabric (reinforcing phase). Three types of resin were used, two types of vinyl ester resin from different manufacturers (HYDREX® 100-LV and Vipel® F013-AAB-00- AOC) and one type of epoxy resin (AIRSTONE® 760E/762H/766H) which were cured by biaxal fiberglass 300 g/m2 . Four laminates (two with epoxy resin (AIRSTONE® 760E/762H/766H) and one with vinyl ester resin - (HYDREX® 100-LV and one with Vipel® F013-AAB-00- AOC) have been prefabricated, each laminate is made in two layers, with a layered resin application and two layers of biaxal fiberglass. After curing, a different type of resin (wet on dry) is applied to each of the laminates as indicated in Table 1. After the first layer of resin, a Fiberglass mat 300g is applied and then again two layers of biaxal fiberglass. As mentioned above, the aim is to combine the lack of shrinkage of the epoxy resin with the better mechanical properties and better productivity of the vinyl ester resin.

After curing, specimens were cut from the adhered laminates to determine shear strength, tensile strength and bending strength. Macrostructural fractographic analysis was carried out at the fracture area of the specimens to analyze the material behavior under mechanical loads.

Table 1. The types of investigated adhesively bonded PMCs

Specimens type	PMC with matrix:	Adhesively bonded with PMC with matrix:
1	Epoxy resin AIRSTONE® 760E/762H/766H	Vinyl ester resin HYDREX® 100-LV
2	Vinyl ester resin HYDREX® 100-LV	Epoxy resin AIRSTONE® 760E/762H/766H
3	Vinyl ester resin Vipel® F013-AAB-00- AOC	Epoxy resin AIRSTONE® 760E/762H/766H
4	Epoxy resin AIRSTONE® 760E/762H/766H	Vinyl ester resin Vipel® F013-AAB-00- AOC

Shear Strength Test Methodology

In order to determine the strength of the adhesively bonded surface of the bonded composite laminates, shear strength testing was conducted by tensile loading according to ASTM D3163 – Standard Test Method for Determining Shear Strength of Adhesively Bonded Rigid Plastic Lap-Shear Joints in Shear by Tension Loading [12]. Three overlap adhesively bonded specimens were fabricated from each type of composite material, the form and size of which are given in Figure 1.

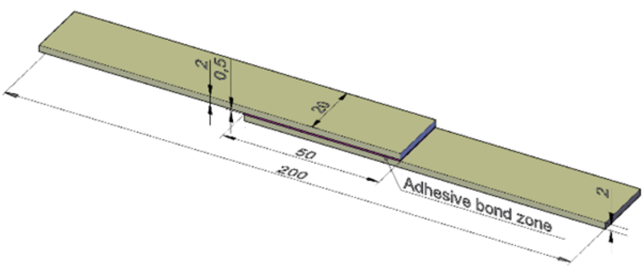

Figure 1. Shear strength test specimen form and size

Tensile Strength Test Methodology

In order to determine the tensile strength of the adhesively bonded composites and to analyze the possibility of delamination in the bond zone, tensile testing was performed according to ISO 527-4:1997

Plastics — Determination of tensile properties — Part 4: "Test conditions for isotropic and orthotropic fibre-reinforced plastic composites" standard [13]. Three specimens were made from each type of bonded composite laminates, the form and size of which are given in figure 2.

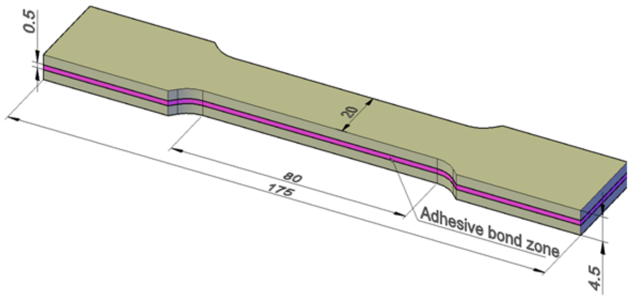

Figure 2. Tensile strength test specimen form and size

Bending Test Methodology

To determine the bending strength and to analyze the material behavior, the bending test was conducted according to ASTM D7264 / D7264M - 21 Standard - Test Method for Flexural Properties of Polymer Matrix Composite Materials [14], which consists of three- point loading with fixed supports and loading nose Figure 3. Again, three specimens were made from each type of adhesively bonded composite laminates with sizes: specimen length l= 140 mm, sample width b = 20 mm and the sample thickness d= 4,5 mm. The support span length L = 70 mm.

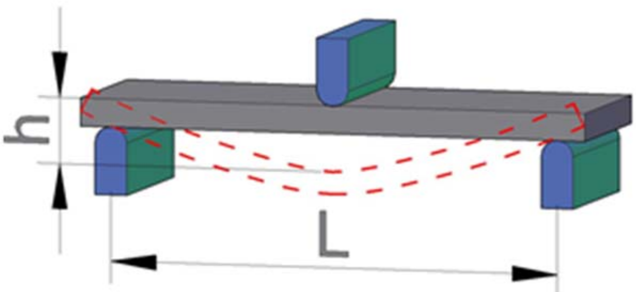

Figure 3. Shem of three-point bending test

The following formula was used to determine the bending strength values:

$$R_{mb} = \frac{3FL}{2bd^2}\ [MPa] \qquad (1)$$

Where: F- load force, N; L- the support span length, mm; b- the sample width, mm; d - the sample thickness, mm;

3. Results and Analyzing

The tests started with shear strength testing under tensile loading conditions, as this test gives the best information about the strength of the adhesively bonded surface and determines the direction of the following studies. As expected, failure of the specimens did not occur due to delamination in the bond zone. From figure 4 it can be seen that the failure was in the boundary region between the adhesively bonded part and the free surface of the laminate, which matrix was made of epoxy resin. This type of failure was observed in all specimens, also all specimens failed in the free surface of the composite with matrix made of epoxy resin, which is due to the lower values of mechanical characteristics of epoxy resin compared to vinyl ester resins. This test proves that the adhesive bonding zone is constructed of a continuous bond based on high cohesive forces, providing the required strength of the PMCs adhesively bonded by this methodology.

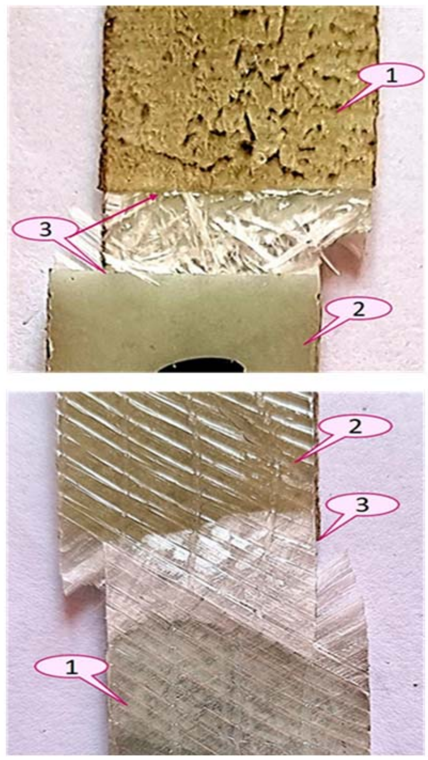

Figure 4. Macrostructure of the fracture surface of the PMCs specimens after shear strength test
1-free surface of the laminate; 2- adhesively bonded part; 3- boundary region;

After the tensile tests, all four types of PMCs were found to have approximately the same tensile strength average values, between 76 MPa and 98 MPa (Table 2). The values obtained are lower than the tensile strength values found in previous studies of similar PMCs with a vinyl ester resin matrix and a bi-axial fiberglass reinforcing phase (125 MPa ÷ 150 MPa) [15], [16]. This difference is due not only to the fact that these composites are of greater thickness (with more layers of the reinforcing phase), but also to the epoxy resin from which the matrix of the laminates of one part of the studied adhesively bonded composites is constructed, since by specification it has lower mechanical characteristics. Once the crack appears due to the applied load, it is a stress concentrator for the second part of the bonded composite with a vinyl ester resin matrix, which promotes its failure and leads to a reduction in the overall strength of the adhesively bonded composite.

Table 2. The results of tensile test

Specimens type	Sample №	*Rm* (MPa)	Average value of *Rm* (MPa)
1	1	82	91
	2	101	
	3	90	
2	1	95	87
	2	88	
	3	89	
3	1	60	78
	2	95	
	3	81	
4	1	97	98
	2	101	
	3	96	

Macrostructural analysis conducted at the fracture area of the studied PMCs revealed that no delamination was observed in the adhesive bonding area (Figure 5). The exception is one of the specimens of the third type of PMCs, where partial delamination was observed in the adhesive bonding area (Figure 5c). Due to the fact that this delamination type was observed in only one specimen out of twelve, it can be concluded that it is most likely due to a confined micro-volume of air (pore ore group of pores) during the bonding of the two laminates making up the composite. These types of defects are stress concentrators which significantly reduce the strength of the material. This explains the significantly lower tensile strength value obtained for this specimen (Table 2), which is also the reason why the average tensile strength value of this type of PMCs is lower. Also, the average tensile strength value of the specimens of the second type of PMCs was lower compared to the others, as one of them failed at a lower load (Table 2).

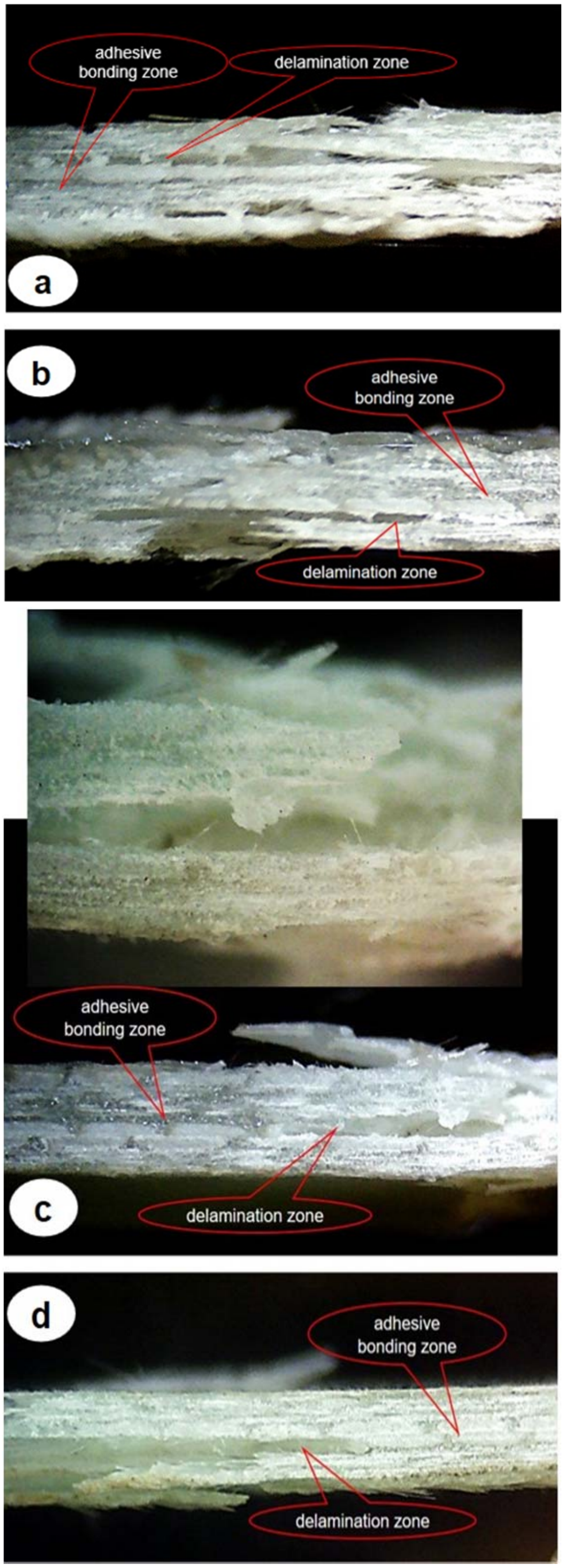

Figure 5. Macrostructure of the fracture surface of the PMCs specimens after tensile strength test: a-1 type PMCs; b- 2 type PMCs; c- type PMCs; d- type PMCs

From the macrostructure of Figure 5d, it can be seen that delamination of larger scale was obtained, but it was outside the bonding area, and most likely again due to a closed micro-volume of air (pore, pore group) during laminate fabrication. It is also evident from the macrostructural analysis carried out Figure 6 that the crack development in the fracture zone is tangential, since the tensile stresses generated pull the fibers out of the matrix, which are at 45° to the tensile force, and form tangential stresses causing the pull of the biaxial layer. This type of failure is characteristic of PMCs with a biaxial reinforcing phase and is associated with the interlacing of the fibers of the biaxial fiberglass at a 45° angle. The result of the tensile test and the macrostructural analysis conducted confirmed that a good adhesive bond with high cohesive strength was formed in the adhesive bonding area of the specimens, which did not affect the overall strength of the adhesively bonded composite, or if there is any influence it is insignificant.

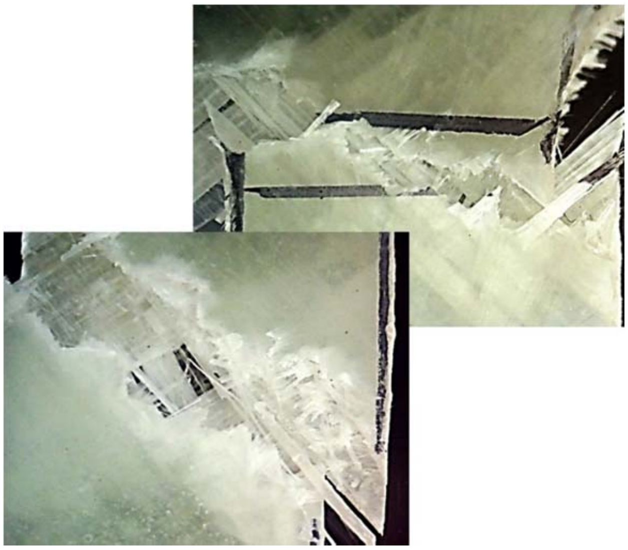

Figure 6. Macrostructure of the front fracture surface of the PMCs specimens after tensile strength test

The bending test results show that all four types of PMCs yielded nearly identical bending strength values, between 179 MPa and 195 MPa (Table 3). The values obtained are similar to the bending strength values found in previous studies of similar PMCs with a vinyl ester resin matrix and a bi-axial fiberglass reinforcing phase (92 MPa ÷ 230 MPa), even exceeding some of them [12], [13]. Again, a large difference was observed in the previous studies due to the difference in the number and location of the curing phase, but it can still be a basis for comparison to some extent. Also, in all four specimens the value of vertical displacement is the same, and vertical displacement is an indicator of flexural deformation, which explains the same values obtained for bending strength. The other conclusion that can be drawn is that due to mechanical loading under bending conditions, there is no difference in the material behavior in the different specimens, i.e. all four types of adhesively bonded PMCs have similar properties.

Table 3. The obtained bending test results

Specimens type	**Specimens №**	***Rmb* (MPa)**	**Average value of *Rmb* (MPa)**	**Vertical displacement *h* (mm)**
1	1	186	186	8,3
	2	190		8,4
	3	183		8,4
2	1	190	179	8,2
	2	165		8,3
	3	183		8,5
3	1	196	195	8,4
	2	191		8,5
	3	197		8,7
4	1	185	185	8,2
	2	191		8,3
	3	180		8,4

From the macro-structural analysis performed, it was found that the fracture at failure zone is typical of this type of composite materials subjected to loading under bending conditions, and no different behavior of the composites studied is observed from the usual (Figure 7a, 7b, 7c, 7d). In all specimens, the material delamination is observed in the outer part of the laminate, which is subjected to tensile stresses, and not in the adhesive bonding area, which is a criterion for good adhesion in the bonding area. From the tensile stresses created in the upper part of the objects studied, local fiber tearing of the reinforcing phase in the failure zone was observed.

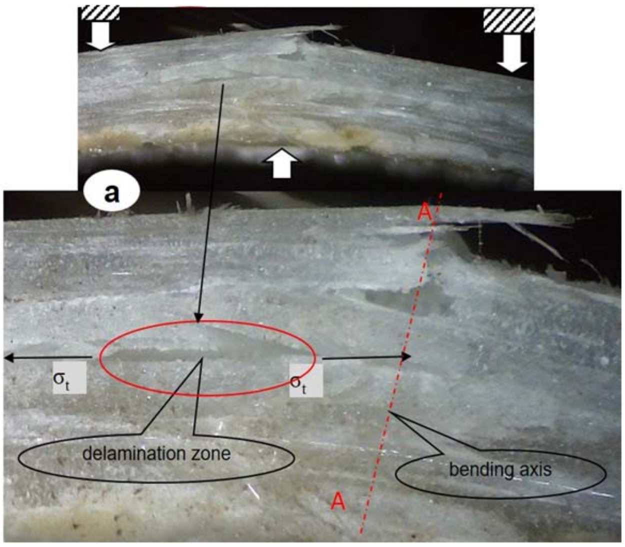

Figure 7a. Macrostructure of the 1st type PMCs specimens after bending strength test

No delamination was observed in the adhesive bonding area provided that the tensile stresses perpendicular to the A-A bending axis created tensile stresses perpendicular to the failure zone (Figure 7 a - 7 d). In some specimens, it can be seen that delamination of a larger scale was obtained, part of which was away from the failure zone and outside the adhesive bonding area (Figure 7 b, 7 c). The reason is that tensile stresses parallel to the bending axis are formed after the surface layer ruptures. The other feature is that, since in biaxial fiberglass the fibers are interlaced in a biaxial direction (+45/-45°), the tensile stresses generated by the bending (perpendicular to the bending axis) pull the fibers located at 45° to the tensile force out of the matrix, thereby generating tangential stresses along the direction of the fibers which displace the knitted layer.

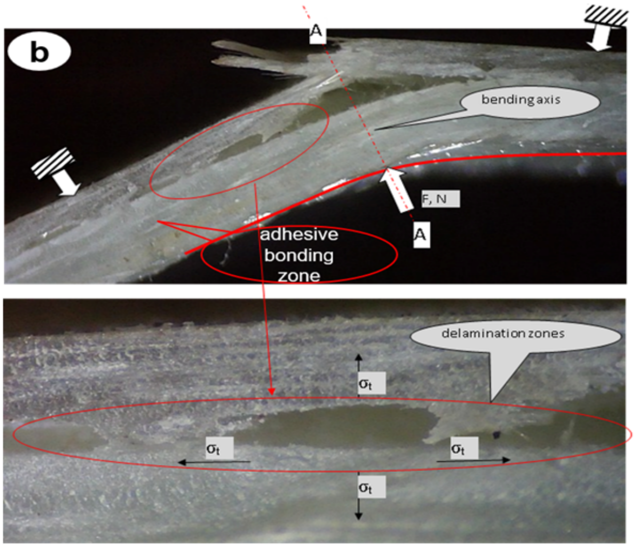

Figure 7b. Macrostructure of the 2nd type PMCs specimens after bending strength test

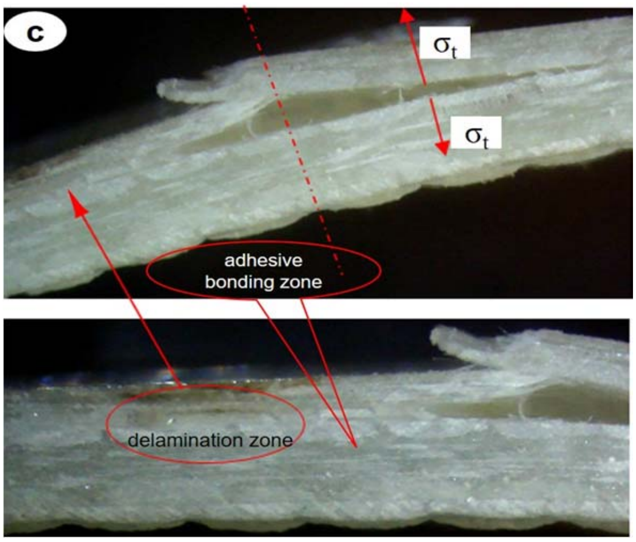

Figure 7c. Macrostructure of the3rd type PMCs specimens after bending strength test

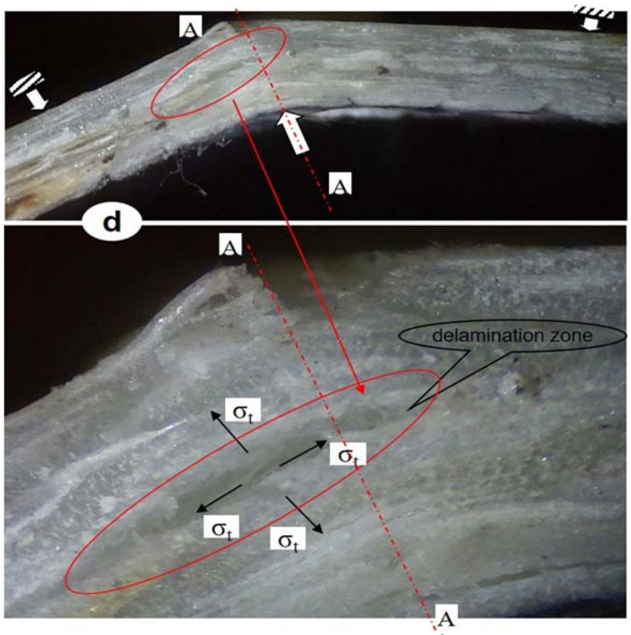

Figure 7d. Macrostructure of the 4th type PMCs specimens after bending strength test

4. Conclusion

After the studies carried out and based on the results obtained, we can conclude that the methodology used for adhesive bonding two types of PMCs with a matrix composed of different (three types of) resins leads to positive results. For all four types of PMCs, almost identical values for tensile strength and bending strength were obtained, similar to the values of tensile strength and bending strength of similar PMCs found in previous studies, indicating that the adhesive bonding area does not affect the mechanical properties of the studied PMCs. Exceptions were observed on a limited number of specimens, where delamination was obtained on a larger scale outside the adhesive bonding area, which was most likely due to closed air micro-volumes (pores, group of pores) during laminate fabrication. These types of defects are stress concentrators that significantly reduce the material strength, but this is generally a problem of the composite laminate manufacturing technology (hand contact) and not of the adhesive bonds during bonding. The results of the mechanical tests and the macrostructural analysis conducted confirmed that a good adhesive bond with high cohesive strength was formed in the adhesive bonding area of the specimens, which did not affect the overall strength of the adhesively bonded composite, or if there is any influence it is insignificant. The behavior of all the bonded composite specimens studied, under mechanical loading conditions, is typical for this type of composite, i.e. all four types of adhesively bonded PMCs have similar mechanical properties.

Finally, we can conclude that the technology for the fabrication of PMCs, by These types of defects are stress concentrators that significantly reduce the material strength, but this is generally a problem of the composite laminate manufacturing technology (hand contact) and not of the adhesive bonds during bonding laminates of PMCs with a matrix of different types of resin, in which vacuum forming and subsequent manual contact building are combined (which can also achieve the fabrication of complex surfaces), leads to positive results that do not alter the properties of the composite.

References

[1]. Bielawski, R., Kowalik, M., Suprynowicz, K., Rządkowski, W., & Pyrzanowski, P. (2016). Investigation of riveted joints of fiberglass composite materials. *Mechanics of Composite Materials*, *52*(2), 199-210.

[2]. Vinson, J. R. (1989). Adhesive bonding of polymer composites. *Polymer Engineering & Science*, *29*(19), 1325-1331.

[3]. Paraschiv, S., Paraschiv, L. S., Serban, A., & Cristea, A. G. (2022). Assessment of onshore wind energy potential under temperate continental climate conditions. *Energy Reports*, *8*, 251-258.

[4]. Serban, A., Paraschiv, L. S., & Paraschiv, S. (2020). Assessment of wind energy potential based on Weibull and Rayleigh distribution models. *Energy Reports*, *6*, 250-267.

[5]. Li, S., Thouless, M. D., Waas, A. M., Schroeder, J. A., & Zavattieri, P. D. (2005). Use of mode-I cohesive-zone models to describe the fracture of an adhesively-bonded polymer-matrix composite. *Composites Science and Technology*, *65*(2), 281-293.

[6]. Skulev, H., & Veselinov, D. (2022, September). The Influence of Substrate Materials on the Microstructure, Microhardness and Wear Resistance of Nickel-base Plasma Spray Coatings. In *Proceedings of the Bulgarian Academy of Sciences,75*(9), 1343-1350.

[7]. Nedelchev, I., Veselinov, D., & Skulev, H. (2019, June). A study of anodizing of Ti-6Al-7Nb alloy. In *2019 16th Conference on Electrical Machines, Drives and Power Systems (ELMA)* (pp. 1-6). IEEE.

[8]. Kadioglu, F. E. R. H. A. T., Avil, E., Ercan, M. E., & Aydogan, T. (2018, May). Effects of different overlap lengths and composite adherend thicknesses on the performance of adhesively-bonded joints under tensile and bending loadings. In *IOP Conference Series: Materials Science and Engineering*, 369(1), 012034. IOP Publishing.

[9]. Hanim, M. A., Tahir, S. M., Jung, D. W., & Calin, R. (2022). Mechanical and physical performance of the advanced biopolymer-based composites with addition of filler and challenges with additive manufacturing. In *Sustainable Biopolymer Composites* (pp. 73-111). Woodhead Publishing.

[10]. Bankova, A. I. (2021). Application of a method for calculating the sizes of perspective objects. In *IOP Conference Series: Materials Science and Engineering, 1031*(1), 012128. IOP Publishing.

[11]. Emmanuel E. Gdoutos (2020). *Fracture Mechanics: An Introduction.* Springer.

[12]. STM D3163 – Standard Test Method for Determining Shear Strength of Adhesively Bonded Rigid Plastic Lap-Shear Joints in Shear by Tension. (2022). Loading. Retrieved from: https://www.ddltesting.com/package-testing/shear-test/ [accessed: 17 August 2022].

[13]. ISO 527-4:1997 Plastics. (2021). Determination of tensile properties — Part 4: Test conditions for isotropic and orthotropic fibre-reinforced plastic composites. Retrieved from: https://www.iso.org/standard/4595.html [accessed: 23 August 2022].

[14]. Standard Test Method for Flexural Properties of Polymer Matrix Composite Materials (2021). Retrieved from: https://www.astm.org/Standards/D7264 [accessed: 01 September 2022].

[15]. Spasova, D., Argirov, Y., Mechkarova, T., & Atanasov, N. (2021, February). Investigation of the suitability of fiber reinforced polymer matrix composites for facilities operating in marine environment. In *IOP Conference Series: Materials Science and Engineering, 1037*(1), 012029. IOP Publishing.

[16]. Spasova, D., Argiro, Y., & Mechkarova, T. (2021). Comparative Analysis of the Mechanical Properties of Polymer Matrix Composites Reinforced with Fiberglass Fabric. *TEM Journal, 10*(4), 1745-1750.