Polish Society of Composite Materials
Polskie Towarzystwo Materiałów Kompozytowych

18: 2 (2018) 103-109

COMPOSITES THEORY AND PRACTICE

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Received (Otrzymano) 17.04.2018

INFLUENCE OF SCALE EFFECT AND TIME ON STRENGTH PROPERTIES OF POLYMER COMPOSITE MADE BY VACUUM METHOD

The paper discusses and attempts to analyze the impact of the method of forming fibrous composite materials on the quality of the laminates. The model assumes that the composite consists of components having individual physico-mechanical properties with a symmetrical structure $[0/90/0/90]_s$. This article uses experimental data of a contact-formed composite (Composite I) and a vacuum bag composite (Composite II) with a polyester matrix (Firestop 8175-w-1) reinforced with E glass matte fabric. Before the samples were cut, the parameters and technological criteria of the formed composite were determined such as the amount of resin and soaking time of the composite reinforcement with the polymer resin. The influence of matrix plastification and a more packed structure in the produced composites on the scale effect (for samples with larger and smaller measuring bases), the dispersion and mean value of strength from the time of aging were determined.

Keywords: polymer, composite, FEM, resin, scale effect

WPŁYW EFEKTU SKALI I CZASU NA WŁAŚCIWOŚCI WYTRZYMAŁOŚCIOWE KOMPOZYTU POLIMEROWEGO WYKONANEGO METODĄ PRÓŻNIOWĄ

Omówiono i podjęto próbę analizy wpływu metody formowania włóknistych materiałów kompozytowych na jakość laminatów. W modelu założono, ze kompozyt składa się z komponentów, mających indywidualne fizykomechaniczne właściwości o symetrycznej strukturze $[0/90/0/90]_s$. W tym artykule wykorzystano eksperymentalne dane kompozytu formowanego metodą kontaktową (kompozyt I) i worka próżniowego (kompozyt II) o osnowie poliestrowej (Firestop 8175-w-1) wzmocnionej mato-tkaniną szklaną typu E. Przed wycięciem próbek określono parametry i kryteria technologiczne formowanego kompozytu: jak ilość i czas nasączania wzmocnienia kompozytu żywicą polimerową. Określono wpływ plastyfikacji osnowy i bardziej upakowanej struktury w wytworzonych kompozytach na efekt skali (dla próbek z większą i mniejszą bazą pomiarową), rozrzut i wartość średniej wytrzymałości od czasu starzenia.

Słowa kluczowe: polimer, kompozyt, MES, żywica, efekt skali

INTRODUCTION

Material properties that relate stress or strain to failure are measured during either a uniaxial test or pure shear test. These properties are referred to collectively as material strengths [1]. Each element of the structure uses real material with specific properties that can change over time. The wide variety of materials used, as well as their behavior in different conditions, actually excludes the possibility of creating a relatively universal description for them [2]. The type of composite testing depends on the state of load of the construction elements and their structure. In the case of thin-walled structures, which the samples are, only the characteristics in the plane of the layer are determined. If the composite intended for the construction is of considerable thickness, it is also possible to conduct tests perpendicular to the layer. When samples from the matrix and reinforcement are made for the tests, the manufacturing technology must be observed to be the same as in the case of manufacturing the element, which guarantees comparable results [3]. Composites with unidirectional fiber are used extremely rarely in the technique. In general, we have a multi-directional fiber arrangement in the composite. Regardless of the technology, these composites can be modeled to be treated as layered composites. The elementary layers of these composites are composites with unidirectional fibers [4].

The testing guidelines are given in the following standards: ISO 14129: 1997 and ASTM D3518. A sample for testing is shown in Figure 1. The composite [± 45°] is made up of unidirectionally reinforced layers or fabrics in which the number of fibers in the warp and weft warp is the same. The width of the sample is in the range of $b = 13 \div 25$ mm. Making samples with a larger width for testing is recommended due to the influence of the edge effect [3].

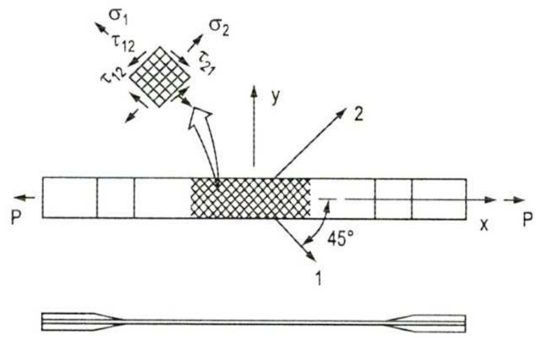

Fig. 1. Sample for shear testing in static tensile test according to ISO 14129:1997 [3]

Rys. 1. Próbka do badań ścinania w statycznej próbie rozciągania wg ISO 14129:1997 [3]

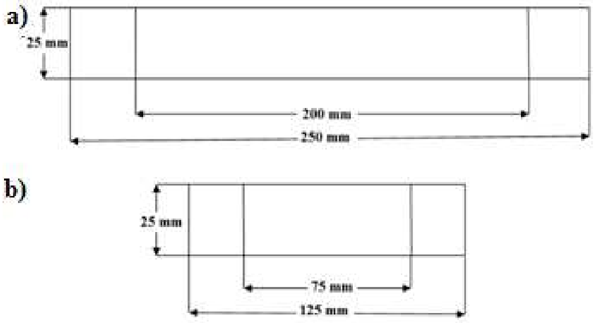

Fig. 2. Geometric dimensions of samples

Rys. 2. Wymiary geometryczne próbek

The statistical theory of fragile strength (scale effect) developed by Waloddi Weibull is based on the assumption that the strength of the entire structure is determined by the strength of its weakest element. According to this theory, the probability of destruction of material $S(\sigma)$ when stresses $\sigma \geq R$ is equal to

$$S(R) = 1 - e^{-Vm(R)} \tag{1}$$

where

$$m(R) = \left(\frac{R}{\sigma_0}\right)^m \tag{2}$$

σ_0 - the minimum possible strength of the material, m - shape parameter, R - strength of the sample.

The dependence of strength on the sample volume according to Weibull's theory is

$$R_{kr} = \frac{A}{v^{\frac{1}{m}}} \tag{3}$$

where: A - the constant depending on the type of material and the state of stress, $\frac{1}{m}$ - the constant for non-homogeneous materials. This leads to the conclusion:

$$\frac{R_2}{R_1} = \left(\frac{v_1}{v_2}\right)^m \tag{4}$$

Weibull's theory has been confirmed in many scientific studies conducted on various types of materials for constructions such as steel and wood [5, 6].

Apart from classic theories and a research experiment, numerical calculations were performed using the finite element method.

The FEM concept assumes that the size (e.g. displacement, stress described by the continuous (primary) function in a given area (continuous fragment of the physical model)) is approximated by a discrete model. The discrete model is composed of a set of continuous functions defined in a finite number of sub-areas called finite elements to which the area in question has been divided [7].

MATERIALS AND METHODS

The authors of this work tend to use samples prepared carefully under controlled conditions, and not under the usual conditions of industrial production. In this way, the occurring processes (cracks in a predetermined part of the tested sample under a load comparable to the load under operating conditions) and phenomena (scale effect) can be controlled, which facilitates interpretation of the obtained results from a given sample. In the case of the fiber forming the composite material intended for the samples, it is necessary to ensure an appropriate fiber content and abide by the manufacturing technology, which should be the same which was used in the production of the material. Defects in the structure significantly affect the quality of the prepared samples and thus the obtained results [8].

In order to carry out the work, two composites: Composites I and II were produced, the first by the contact method and Composite II by one of the vacuum methods, which include the vacuum bag method. It is a combination of manual lamination (the composite reinforcement is infiltrated as in the case of manual lamination) and vacuum lamination. The reinforcement in the form of biaxial from glass fibers with the basis weight of 450 g/cm^2 was used and the matrix in the form of a resin from the BÜFA company.

TABLE 1. **Properties of Firestop 8175-w-1resin, BÜFA company [9]**

TABELA 1. **Właściwości żywicy Firestop 8175-w-1 firmy BÜFA [9]**

Property	Test method	Value	Unit
Density at 20°C	DIN 53 217/2	1.25÷1.35	[g/ml]
Viscosity at 20°C	ISO 2555	20000÷25000	[mPas]
Monomer content		18÷20	[%]
Flash point	DIN 53 213	29	[°C]

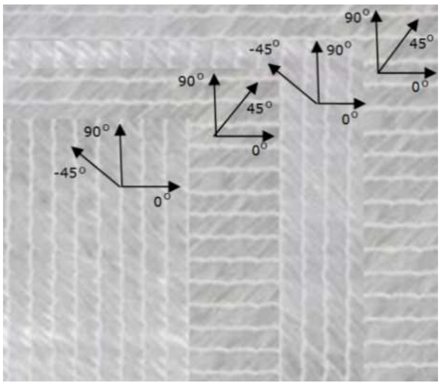

Fig. 3. Diagram of layering composite of Biaxial material $[-45/45/-45/45]_n$

Rys. 3. Schemat ułożenia warstw kompozytu z materiału typu Biaxial $[-45/45/-45/45]_n$

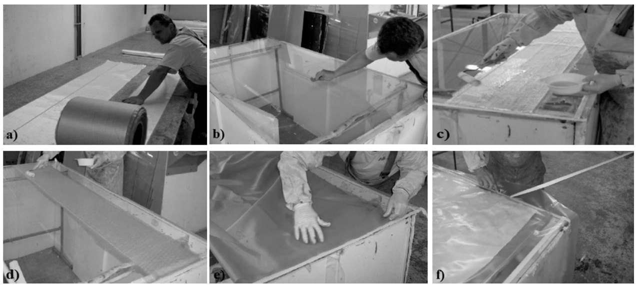

Fig. 4. Forming stages of Composite material II made by vacuum bag method: a) cutting out and preparing reinforcement; b) mold surface preparation; c) infiltrating individual laminated reinforcement layers with resin; d) ready composite material II; e) application of auxiliary layers under film such as peel ply, breathing material; f) application of double-sided sealing tape made of special bonded film around mold edge, under which composite material components (and auxiliary material) are present [10]

Rys. 4. Etapy formowania materiału kompozytowego II metodą worka próżniowego: a) wycinanie i przygotowanie wzmocnienia; b) przygotowanie powierzchni formy; c) przesączanie poszczególnych warstw laminatu żywicą; przesączony materiał II; e) nakładanie tkaniny delaminacyjnej; f) montaż worka próżniowego za pomocą taśmy dwustronnej [10]

In the numerical part of the analysis, the ABAQUS program was used. ABAQUS is a package of programs used to solve complex engineering problems. ABAQUS is widely used in the world in the machine and automotive, metallurgical and mining, shipbuilding and aviation industries - wherever reliable strength assessment of machine elements or engineering structures is necessary. What distinguishes ABAQUS from other programs is its modular construction. It allows the possibility of creating combinations of any of their elements. The user can create any combination of finite elements, materials, analysis procedures and load sequences. The library of analysis procedures offers the possibility to analyze the state of stresses, strains and displacements of arbitrarily non-linear processes and also established and unsteady heat flow, mass transport, acoustic and piezoelectric analyses. These analyses can be carried out independently, sequentially or fully coupled with stress analysis [11].

RESULTS ANALYSIS

The samples were subjected to a static tensile test (according to ISO 14129: 1997) with a stretching speed of 1 and 2 mm/min (respectively Composite I and Composite II) on the universal testing machine Instron 8501 in the Materials Strength Laboratory - Laser Technology Center of Metals of Kielce University of Technology.

The following designations of samples subjected to a static tensile strength test were used:

- "POLY1_1-xx" and "POLY2_2-xx": where, 'POLY1' and 'POLY2' means, respectively, - samples with a measuring base of 150 mm, Composite I and Composite II sample series, with cutting angles of: 1 - 0°; 2 - 90°, along with following number 'xx';
- "POLY3_1-xx" and "POLY4_2-xx": where, 'POLY3' and 'POLY4' means, respectively, - samples with a measuring base of 60 mm, Composite I and Composite II sample series, with cutting angles of: 1 - 0°; 2 - 90°, along with following sample number 'xx';
- "POLY14_1-xx" and "POLY24_2-xx": where, 'POLY1' and 'POLY2' means, respectively, - samples with a measuring base 150 mm, Composite I and Composite II sample series, with cutting angles of: 1 - 0°; 2 - 90°, along with following sample number 'xx' after four years of exposure;
- "POLY34_1-xx" and "POLY44_2-xx": where, 'POLY3' and 'POLY4' means respectively, - samples with a 60 mm measuring base, Composite I and Composite II sample series, with cutting angles of: 1 - 0°; 2 - 90°, along with following sample number 'xx' after four years of exposure;.

The test results obtained from the static tensile strength test of Composite I sample series (Fig. 5, Table 2) had a larger dispersion of strength values in comparison to the Composite II sample series, formed by the vacuum bag method (Fig. 6; Table 3) with an average strength of 5÷12 and 6÷8% respectively (for samples cut along the reinforcement).

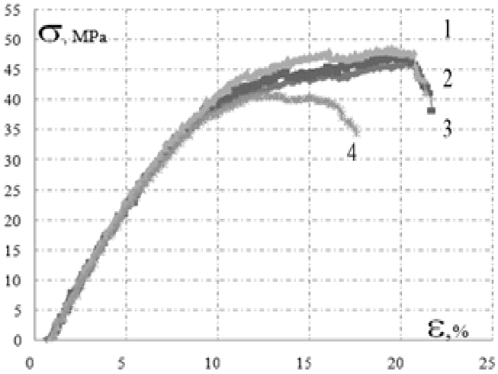

Fig. 5. Composite I σ-ε curves for samples with L_{BP} = 150 mm: 1 - sample POLY1_2-1; 2 - sample POLY1_2-2; 3 - sample POLY1_2-3; 4 - sample POLY1_2-4

Rys. 5. Krzywe σ-ε kompozytu I dla próbek z L_{BP} = 150 mm: 1 - próbka POLY1_2-1; 2 - próbka POLY1_2-2; 3 - próbka POLY1_2-3; 4 - próbka POLY1_2-4

TABLE 2. **List of mean strength of Composite I sample series, with cutting angle of 90° and measuring base of 150 mm (after static tensile strength test)**

TABELA 2. **Zestawienie średniej wytrzymałości kompozytu I ciętego pod kątem 90° z bazą pomiarowa 150 mm (po statycznej próbie rozciągania)**

Sample series	Strength σ [MPa]	Young's modulus E [GPa]	Deformation ε [%]	Dispersion σ [%]
POLY1_2-1	48.0	2.9	21.0	5÷12
POLY1_2-2	46.3	2.6	21.0	
POLY1_2-3	45.0	2.5	21.1	
POLY1_2-4	40.4	2.3	16.5	
Mean	**45.6**	**26**	**20.0**	

The defects caused at the production stage, worsened the mechanical properties of the formed laminates (as a result of manufacturing technological errors, or other secondary operations such as sample preparation). With an increase in the angle of arrangement from 0 to 90°, the mean strength of the Composite I and Composite II sample series, were improved by 9 and 15%, respectively.

TABLE 3. **List of mean strength of Composite II sample series with cutting angle of 90° and 150 mm measuring base (after static tensile strength test)**

TABELA 3. **Zestawienie średniej wytrzymałości kompozytu II ciętego pod kątem 90° z bazą pomiarowa 150 mm (po statycznej próbie rozciągania)**

Sample series	Strength σ [MPa]	Young's modulus E [GPa]	Deformation ε [%]	Dispersion σ [%]
POLY2_2-1	89.4	4.9	21.0	6÷8
POLY2_2-2	86.0	4.8	19.0	
POLY2_2-3	78.0	4.4	18.4	
POLY2_2-4	83.9	4.7	17.0	
Mean	**84.3**	**4.7**	**18.9**	

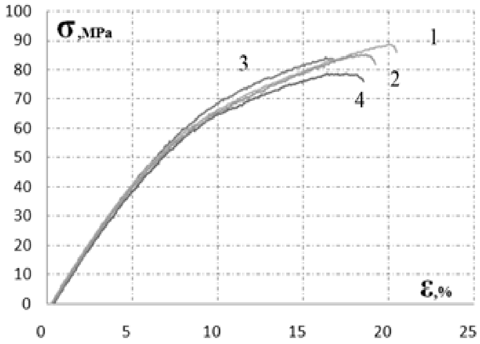

Fig. 6. Composite II σ-ε curves for samples with L_{BP} = 150 mm: 1- sample POLY2_2-1; 2 - sample POLY2_2-2; 3 - sample POLY2_2-3; 4 - sample POLY2_2-4

Rys. 6. Krzywe σ-ε kompozytu II dla próbek z L_{BP} = 150 mm: 1 - próbka POLY2_2-1; 2 - próbka POLY2_2-2; 3 - próbka POLY2_2-3; 4 - próbka POLY2_2-4

The highest mean strength for the 2-directional Composite II sample series, with the load acting at the angle of 90° relative to the reinforcement, was 89.4 MPa (a 2-directional Composite I sample series also had the above relationship which means σ was about 0.4% lower than the Composite II sample series). On the other hand, the determined dispersions and mean strength values, obtained from 4 samples, were used to estimate the mean strength of the Composite II sample series, cut not only along and perpendicular to the reinforcement, but also at the angle of 45° (Table 4).

TABLE 4. **Comparison of mean strength of Composite I and II sample series, cut at angle of 45° with 150 mm measuring base (after static tensile strength test)**

TABELA 4. **Zestawienie średniej wytrzymałości kompozytu I i II ciętego pod kątem 45° z bazą pomiarowa 150 mm (po statycznej próbie rozciągania)**

Dispersion properties/composite	II	I
σ [MPa]	674÷782	240÷70,0
E [GPa]	87÷102	9.20÷9.9
Mean σ [MPa]	**737**	**58.2**
Mean E [GPa]	**9.3**	**9.5**

The tests carried out for the Composite I and Composite II sample series, with a smaller measuring base (Figs. 7 and 8), retained the above relationships, as found for samples with a larger measuring base. Therewith, they had a higher mean strength of 36% (68.6 MPa) and 13% (97.6 MPa) respectively, significantly reducing the dispersion of strength, with respect to the longer measuring base.

The observed deterioration of strength of the samples with the longer measuring length base is explained by the higher probability of defect occurrence than in the case of samples with a smaller measuring base, produced with a different method. The process of delamination is mostly influenced by normal inter-layer (S) and tangential (τ) stresses, a sign of which is "swelling" of the free edge. This phenomenon is very sensitive to the quality and imperfections of the composite structure produced in the material forming process [12, 13]. In recent years, understanding of the boundary effect focuses rather on the depicting this phenomenon than on creating methods allowing its impact on the strength of complex materials, or structural elements to be taken into account. Stratification at the edges of the samples, or the scale effect, becomes more visible when the composite structure is destroyed in dynamic studies, and statistical analysis allows one to more accurately take into account these phenomena caused by technological defects and the following destruction of the material.

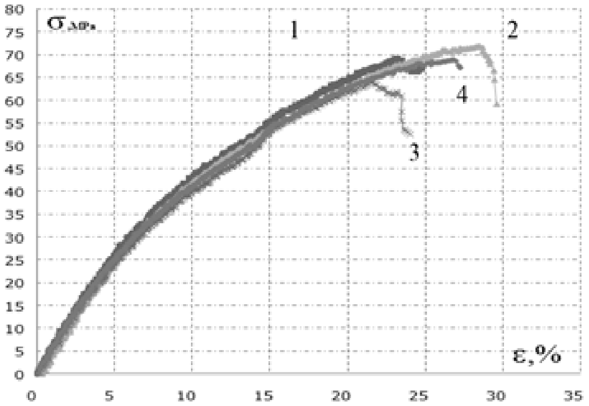

Fig. 7. Composite I σ-ε curves for samples with L_{BP} = 60 mm: 1 - POLY3_2-1; 2 - sample POLY3_2-2; 3 - sample POLY3_2-3; 4 - sample POLY3_2-4

Rys. 7. Krzywe σ-ε kompozytu I dla próbek z L_{BP} = 60 mm: 1 - próbka POLY3_2-1; 2 - próbka POLY3_2-2; 3 - próbka POLY3_2-3; 4 - próbka POLY3_2-4

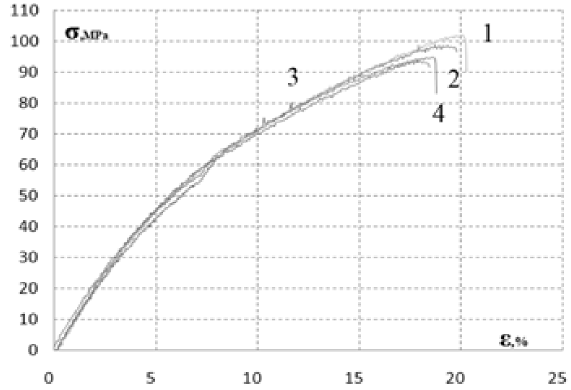

Fig. 8. Composite II σ-ε curves for samples with L_{BP} = 60 mm: 1 - POLY4_2-1 sample; 2 - sample POLY4_2-2; 3 - POLY4_2-3; 4 - POLY4_2-4 sample

Rys. 8. Krzywe σ-ε kompozytu II dla próbek z L_{BP} = 60 mm: 1 - próbka POLY4_2-1; 2 - próbka POLY4_2-2; 3 - próbka POLY4_2-3; 4 - próbka POLY4_2-4

The instability of changes in the strength characteristics of Composite I (formed by the contact method), does not guarantee reproducibility of results, which is not observed when determining the strength of samples made with the vacuum bag method (Composite II sample series). After four years of exposure (influence of time, after aging), the Composite I sample series (Fig. 9) and from Composite II samples (Fig. 10), had improved mean strength results (Table 5) by 90 and 5% respectively (27 and 4%) with the measuring base L_{BP} = 150 mm (L_{BP} = 60 mm). A change in the nature of the dependence for σ-ε and lower values of yield limits as a result of matrix plasticization are observed. Destruction of the material may be the result of overlapping effects, related to the quality and technological parameters in the production state of polymer composites. We observe the same effect for a smaller measuring base (Fig. 11 and 12).

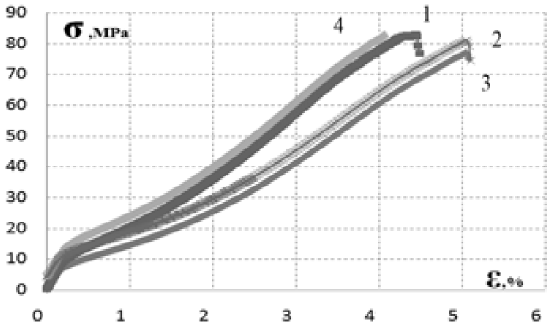

Fig. 9. Composite I σ-ε curves for samples with L_{BP} = 150 mm: 1 - sample POLY14_2-1; 2 - sample POLY14_2-2; 3 - sample POLY14_2-3; 4 - sample POLY14_2-4

Rys. 9. Krzywe σ-ε kompozytu I dla próbek z L_{BP} = 150 mm: 1 - próbka POLY14_2-1; 2 - próbka POLY14_2-2; 3 - próbka POLY14_2-3; 4 - próbka POLY14_2-4

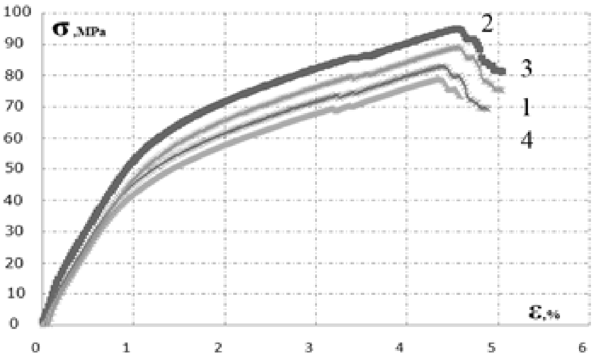

Fig. 10. Composite II σ-ε curves for samples with L_{BP} = 150 mm: 1 - sample POLY24_2-1; 2 - sample POLY24_2-2; 3 - sample POLY24_2-3; 4 - sample POLY24_2-4

Rys. 10. Krzywe σ-ε kompozytu II dla próbek z L_{BP} = 150 mm: 1 - próbka POLY24_2-1; 2 - próbka POLY24_2-2; 3 - próbka POLY24_2-3; 4 - próbka POLY24_2-4

TABLE 5. **List of mechanical properties of Composite I and II sample series, with cutting angle of 90° and various base lengths, after 4 years of exposure**

TABELA 5. **Zestawienie właściwości mechanicznych kompozytów I i II ciętego pod kątem 90° z różną bazą pomiarową po 4 latach ekspozycji**

Samples of composite/ properties	Strength σ [MPa]	Young's modulus E [GPa]	Force F [kN]	Dispersion σ [%]
I (L_{BP} = 150 mm)	81.1	1.829	5.816	7.8
I (L_{BP} = 60 mm)	93.7	1.256	6.755	10.0
II (L_{BP} = 150 mm)	86.0	1.08	2.713	11.0
II (L_{BP} = 60 mm)	101.2	1.706	3.203	5.0

The accumulation of defects and damages in the analyzed structures (Fig. 13), results in the loss of adhesion at the interface between the fiber and the matrix (so-called debonding) - is microstructural damage occurring in the earliest stages of the degradation process of polymer composites. At this stage of material destruction, adhesive cracks do not exert a significant influence on the macroscopic material characteristics, but are, as usual, the first stage of the material destruction process. Damage to the boundary layer arises as a result of exceeding the values of critical stresses, i.e. normal stresses to the lateral surface of the fiber, and to the tangential surface of the fiber (caused by shearing deformation of the contact zone between the fiber and the warp).

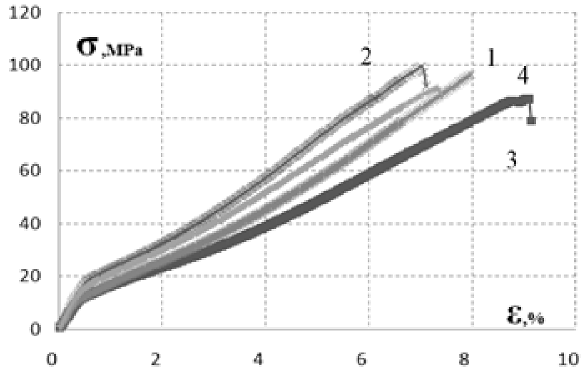

Fig. 11. Composite I σ-ε curves for samples with L_{BP} = 60mm: 1 - sample POLY34_2-1; 2 - sample POLY34_2-2; 3 - sample POLY34_2-3; 4 - sample POLY34_2-4

Rys. 11. Krzywe σ-ε kompozytu I dla próbek z L_{BP} = 60 mm: 1 - próbka POLY34_2-1; 2 - próbka POLY34_2-2; 3 - próbka POLY34_2-3; 4 - próbka POLY34_2-4

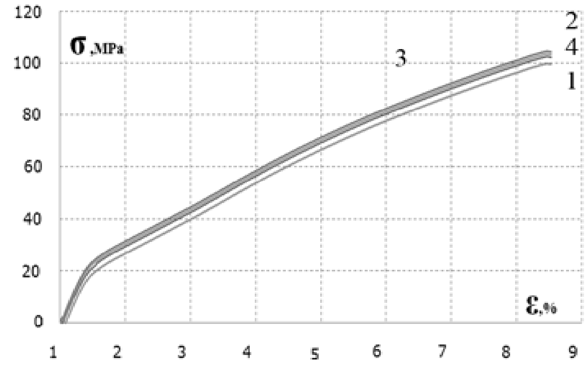

Fig. 12. Composite II σ-ε curves for samples with L_{BP} = 60 mm: 1 - sample POLY44_2-1; 2 - sample POLY44_2-2; 3 - sample POLY44_2-3; 4 - sample POLY44_2-4

Rys. 12. Krzywe σ-ε kompozytu II dla próbek z L_{BP} = 60 mm: 1 - próbka POLY44_2-1; 2 - próbka POLY44_2-2; 3 - próbka POLY44_2-3; 4 - próbka POLY44_2-4

Those defects (Fig. 13) most often appear on the edges of finished elements, introduced by operations (e.g. cutting), or as a result of failure to maintain curing times and a heterogeneous packing structure and an insufficient ratio of resin among successive mat layers (especially in Composite I - Fig. 14). The laminate (Composite II sample series), produced by the vacuum bag method, has a uniform and 20% smaller thickness than Composite I (formed by the contact method [14]).

It results from the course of the technological process, uneven (in the whole volume) fiber distribution, local fiber discontinuities, lack of adhesion at the fiber-matrix boundary, as well as imperfections in the matrices, in the form of voids, microcracks or gaps.

All the defects and phenomena (developed during the technological process) degrade the mechanical properties of the material as well as the aesthetic qualities of the product. These phenomena are very sensitive to the quality and imperfections of the FMC structure created due to the material forming process. Research of damage propagation in laminates is usually based on observations and experimental measurements. Equations in the field of fracture mechanics allow the modeling of cracking and delamination processes in the linear-elastic range or elastic-plastic lag and do not take into account the visco-elastic phenomena occurring during these processes. FEM is effective in modeling the cracking and delamination of polymeric laminates [15].

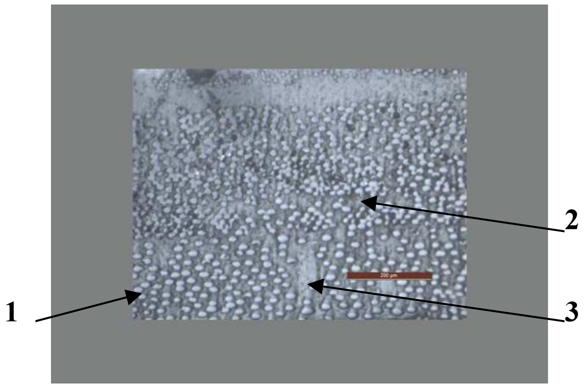

Fig. 13. Microstructure, showing defects in form of: 1 - micro-gaps; 2 - micro cracks of components; 3 - voids in polyester matrix

Rys. 13. Mikrostruktury z defektami w postaci: 1 - mikroszczeliny; 2 - mikropęknięcia komponentów; 3 - pustki w osnowie poliestrowej

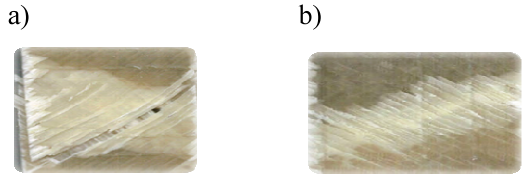

Fig. 14. Breakthrough of composite samples from Composite I (a) and Composite II (b)

Rys. 14. Przełom próbki kompozytu I (a) i kompozytu II (b)

Based on the experimental data, two models were made using the ABAQUS program (Figs 15 and 16). The ABAQUS program has a modular construction; thanks to the composite layup manager tab located in the Property module, it is possible to arrange the fabric at specific angles (in this case [45/45/–45/45]). The finite element method suggests that a composite material crack (Figs. 15a and 16a) just at the beginning of the measuring base of its lower or upper part applies to both long samples (dimensions: Fig. 2a) and short ones (Fig. 2b). However, the largest displacements in the unsymmetrical composition with the arrangement $[-45/45/-45/45]_n$ are on the right side of the sample and are presented in the form of triangles separated from each other by strands with lower stresses. It should be remembered that the finite element method does not take into account material defects such as microspheres and microcracks formed in the laminate as a result of resin gelation. Due to the anisotropy of the material (Fig. 16a and b), they represent displacements in the direction of the axis in accordance with the angle of material orientation, in this case it is the x-axis.

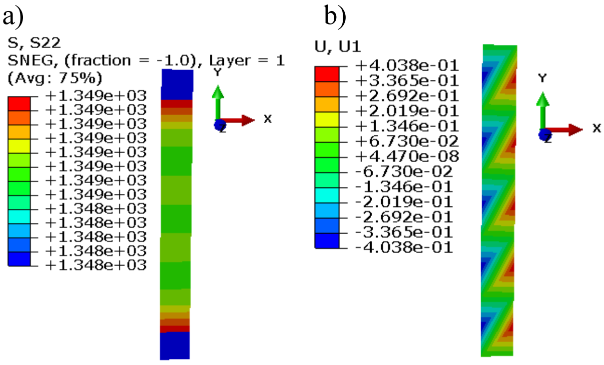

Fig. 15. Tensile stress (dimensions Fig. 2a) (a) and displacement (dimensions Fig. 2a) (b)

Rys. 15. Naprężenia rozciągające (wymiary rys.2a) (a) i przemieszczenie (wymiary rys.2a) (b)

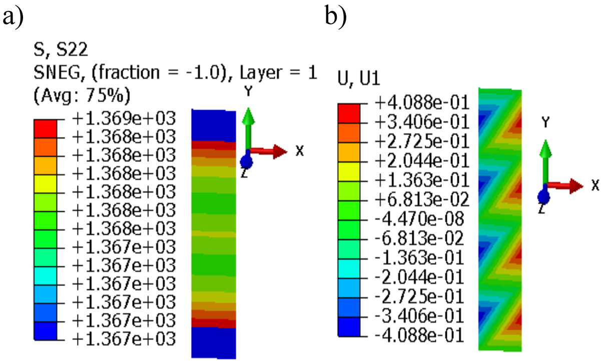

Fig 16. Tensile stress (dimensions Fig. 2b) (a) and displacement (dimensions Fig. 2b) (b)

Rys. 16. Naprężenia rozciągające (wymiary rys.2b) (a) i przemieszczenie (wymiary rys.2b) (b)

CONCLUSIONS

The scientific literature contains very little information about the influence of the scale effect on the elastic and plastic properties of the material. This is due to the complexity of the experiments and the difficulty of interpreting the results. In a composite, i.e. a linear-elastic or visco-elastic material, complicated stress distribution processes take place, the values of which depend on the heterogeneous structure of the composite and the type of defects in it (microspheres and microcracks).

By analyzing the composite (Fig. 3) subjected to a static tensile test, a high elasticity of materials amounting to approx. 20% can be observed. After 4 years, the elasticity decreases to about 8%. With time, the elasticity decreases and with it the strength increases.

The resulting phenomena are caused by the occurrence of technological defects and the choice of the type of matrix in the form of a resin with adequate reactivity. Samples produced by the vacuum bag method (Fig. 4) are characterized by a higher strength of about 30÷46% respectively for samples l = 150 mm and l = 60 mm hand lay-up samples. A greater spread of strength was observed for long L = 150 mm (short: L = 60mm) samples by 12 and 18% (8.5 and 11.5%) made by the vacuum bag method (Composite II), respectively, than the contact method (Composite I). Despite the variety of composite techniques and various defects associated with this method, the scale effect occurred in all the tested samples (as a result of matrix plasticization).

The above analyses require further verification work, taking into account the quality and architecture of stacking the layers in the material.

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