Experimental research and statistic analysis of polymer composite adhesive joints strength

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Abstract. The aim of this paper is to determine the effect of arrangement of fibreglass fabric plies in a polymer composite on adhesive joint strength. Based on the experimental results, the real effect of plies arrangement and their most favourable configuration with respect to strength is determined. The experiments were performed on 3 types of composites which had different fibre orientations. The composites had three plies of fabric. The plies arrangement in Composite I was unchanged, in Composite II the central ply had the 45° orientation, while in Composite III the outside ply (tangential to the adhesive layer) was oriented at 45°. Composite plates were first cut into smaller specimens and then adhesive-bonded in different combinations with Epidian 61/Z1/100:10 epoxy adhesive. After stabilizing, the single-lap adhesive joints were subjected to shear strength tests. It was noted that plies arrangement in composite materials affects the strength of adhesive joints made of these composites between the values of the strength of the confidence level of 0.95. The statistical analysis of the results also showed that there are no statistical significant differences in average values of surface free energy (0.95 confidence level).

1. Introduction
Currently, composite materials constitute a wide and rapidly developing area of polymers. Composites are mainly used in the production of aircraft, sports equipment, and different types of boats and tanks [1,2] as well as pipelines [3,4] and their fittings and also bearings [5] and tubular structures [6,7]. Products made of materials such as fibreglass-reinforced polymers play a significant role in industry [4]. These components include fillers and polymer matrix. The filler material can be made fibres, fabric, paper, and others [8-11]. The polymer matrix can be made of resins, such as epoxy resins, polyester resins, phenol-formaldehyde resins or maleic resins [12-14]. One of the methods of composite forming is laminating [3,15]. Laminates of fabrics exhibit the warp and weft maximum mechanical properties, while at 45° to the directions of these characteristics are much worse [12]. The textile fiber roving strands are interleaved in parallel. This allows obtaining a particularly high strength bidirectional, with a total use of considerable strength roving bands of parallel fibers. In order to achieve a high strength uni-directional in the plane is used as reinforcement fabric unidirectional (targeted), arranged in parallel layers [16]. The various laminates, which have various the
arrangements of plies and amount of plies, were tested. Katman et al. [17] tested the laminate thickness of 2 mm which was targeted using 16 plies (each ply is approximately 0.125 mm thick) with (01/90)\textsubscript{6} lay-up. Bénard et al. [12] investigated the composite samples which are produced, respectively, with 8 and 10 pre-impregnate plies of carbon and glass fibres in the same orientation. There are generally two kinds of joining methods for composite structures [4,18,19]: (1) mechanical fastening and (2) adhesive bonding. Adhesive bonding is an alternative to conventional joining methods in structural applications, and it provides many advantages such as lower structural weight, improved damage tolerance and design flexibility over mechanical fasteners [6,8,17,20]. This paper investigates the effect of arrangement of individual plies of fibreglass fabric in a composite material on the strength of adhesive joints. On the basis of the analysis of the studies specified the actual impact of the arrangement of layers and the best of their arrangement affect strength of adhesive joints.

2. Experimental test

2.1. Characteristics of the tested composites

Three polymer composites made of two constituent materials were fabricated. The reinforcement material was plain weave fibreglass fabric 200g/m\textsuperscript{2} (figure 1), while the matrix material was POLIMAL 1094 polyester resin. The schematic of plain weave fibreglass fabric was presented in figure 2.

![Figure 1. View of fibreglass fabric 200g/m\textsuperscript{2}](image1)

**Figure 2.** View of plain weave scheme of a rare woven.

| Name of material | Plies arrangement |
|------------------|-------------------|
| Composite (I): plain weave fibreglass fabric 200g/m\textsuperscript{2}, Matrix - polyester, 3 plies | 0°/0°/0° |
| Composite (II): plain weave fibreglass fabric 200g/m\textsuperscript{2}, Matrix - polyester, 3 plies | 0°/45°/0° |
| Composite (III): plain weave fibreglass fabric 200g/m\textsuperscript{2}, Matrix - polyester, 3 plies | 45°/0°/0° |

The schematic of plain weave fibreglass fabric was presented in figure 2. In plain weave, each warp thread is interwoven and intersects with each weft thread and thus the fabric is tightly bound and rigid. The composites had three plies of fabric. The plies arrangement in Composite I was unchanged, in Composite II the central ply had the 45° orientation, while in Composite III the outside ply (tangential to the adhesive layer) was oriented at 45° (figures 3 – 6 and table 1).

Prior to fabricating each composite material, fibreglass fabric and polyester resin (POLIMAL 1094, produced by Organika Sarzyna, Poland) were mixed with Metox-50 curing agent. The resin and curing agent were thoroughly mixed in a ratio of 100:10. Polimal 1094 is orthophthalic structural resin, medium-flexible, accelerated and low emission of styrene. Metox-50 is a peroxide initiator for copolymerization of unsaturated polyester resins and vinyl ester. This hardener contains methyl ethyl ketone peroxide and it is 20% solution in dimethylphthalate.
The three prepared composite materials were left to cure for 4 days. After that period, each composite was removed from the mould, cut and numbered. The samples were cut by a FNU-50E milling machine. The cut composites were used four blade cutter mill with skew cuts in the working part of the CVD diamond coating for the assay JRT0600 a diameter of 6 mm. The cut was implemented with the following technological parameters: RPM 9000 min$^{-1}$ and a feed of 1000 mm/min. Individual samples were numbered was to facilitate experimental tests and detection of defects.

2.2. Adhesive joints

The experiments were performed on samples produced in compliance with the above procedure. Each sample had a length of 100 mm and a width of 25 mm and a thickness of 2 mm. Based on the combinations with repetitions (1), it was calculated that the 3 types of composites combined using two constituent materials can produce samples with 6 variants of adhesive joints combinations, as listed in table 2. Calculated from the formula ([21]) (2):

$$C^n_k = \frac{(k + n - 1)!}{k!(n-1)!}$$  \hspace{1cm} (1)

$k = 2 - k$, are the combinations of elements with repetitions, $n = 3 - n$, is the number of elements in the set.

Adhesive joints of polymer composite were made using an epoxy adhesive made from Epidian 6 epoxy resin and triethylenetetraamine (produced by Organika Sarzyna, Poland) curing agent which were mechanically mixed in a weight ratio of 100:10, taking into account different combinations of types of composite materials listed in table 2. The lap joint length was 15 mm and the adhesive thickness (tad) is equal 0.14±0.08 mm. The some configuration of single lap adhesive joints of polymer composites is presented in figures 7 – 9 (red plie had the 45$^\circ$ orientation).
Table 2. Combinations of composite adhesive joints.

| Type of composite | Composite (I) | Composite (II) | Composite (III) |
|-------------------|--------------|----------------|-----------------|
| Composite (I)     | I-I          |                |                 |
| Composite (II)    | I-II         | II-II          |                 |
| Composite (III)   | I-III        | II-III         | III-III         |

Prior to adhesive bonding, the samples were degreased with Loctite 7063, an acetone-based degreasing agent. The procedure of degreasing was presented in [22]. The adhesive joints were made using Epidian 6/Z1/100:10 epoxy adhesive which was subjected to single-stage curing for 168 hours in an ambient temperature of 23-25°C, under a load of 0.0016 MPa and a humidity of 22-31%.

The strength tests were performed on shear-loaded single-lap polymer composite adhesive joints (figure 10) in compliance with the standard DIN EN 1465 and the test speed was 5 mm/min. The tests were performed on 10-12 test pieces per each batch.

2.3. Statistical analysis method

In order to proceed to the analysis of the test results, some parameters of descriptive statistics should be determined, which sets certain characteristics describing the properties of the tested characteristics [21]. Evaluation of distribution by means of these measures may include the position of characteristics, their diversity, symmetry of distribution and concentration. The normality of the distribution can be determined by analysing the above mentioned parameters and using the Shapiro-Wilk test, which is considered the best test to check the normality of distribution of a random variable of a small sample. The main advantage of this test is its high power, which means that for a fixed confidence level \( \alpha \), the probability of rejecting the null hypothesis \( H_0 \) when it is false, is greater than for the other tests of this type. In order to check the veracity of the hypotheses posed in relation to the test results, the F test (Fisher Snedecor test) should be used, through which it is possible to verify the hypothesis of equality of two population’s variances. If the hypothesis is not rejected, then the t-Student test should be used, and if the hypothesis is rejected, then the Cochran-Cox test should be used [21]. The development of the data involved tests of significance to compare the mean values of the tested trait in two general populations. The test included the first type error, i.e. rejection of the tested
hypothesis if it is true. The first type error is called the level of significance and was assumed at $\alpha = 0.05$. The samples are to be treated as small due to the fact that the set included up to 30 elements.

2.4. Preparation for experiments: criteria for comparison

The adhesive joints of polymer composites were divided into 4 groups. In each of these groups the results were investigated with respect to a different set of comparative criteria.

**Group 1** – the joints in Group 1 were compared with regard to the effect of change (shifting the turned ply) relative to the ply in Composite I that was constant in terms of its arrangement (table 3).

**Group 2** – in this group, it was assumed that the plies arrangement on one of the sides was constant and had the form of $0^\circ/45^\circ/0^\circ$ (Composite II), while it was changed on the other side of the adhesive joint (table 4).

**Group 3** – the results in Group 3 were examined with respect to the effect of shifting the turned ply on the one side of the adhesive layer, while the plies arrangement was constant on the other side and had the form of $0^\circ/0^\circ/45^\circ$, the turned ply was located the closest to the adhesive layer (table 5).

**Group 4** – the results in Group 6 were examined with respect to the effect of symmetry change in plies arrangement on the strength of adhesive joints (table 6).

**Table 3. Adhesive joints – Group 1.**

| Composite plies arrangement | Composite plies arrangement | Type of composite adhesive joint |
|-----------------------------|-----------------------------|---------------------------------|
| $0^\circ/0^\circ/0^\circ$   | $0^\circ/0^\circ/0^\circ$   | I-I                              |
| $0^\circ/0^\circ/0^\circ$   | $0^\circ/45^\circ/0^\circ$  | I-II                             |
| $0^\circ/0^\circ/0^\circ$   | $45^\circ/0^\circ/0^\circ$  | I-III                            |

**Table 4. Adhesive joints – Group 2.**

| Composite plies arrangement | Composite plies arrangement | Type of composite adhesive joint |
|-----------------------------|-----------------------------|---------------------------------|
| $0^\circ/45^\circ/0^\circ$  | $0^\circ/0^\circ/0^\circ$   | II-I                             |
| $0^\circ/45^\circ/0^\circ$  | $0^\circ/45^\circ/0^\circ$  | II-II                            |
| $0^\circ/45^\circ/0^\circ$  | $45^\circ/0^\circ/0^\circ$  | II-III                           |

**Table 5. Adhesive joints – Group 3.**

| Composite plies arrangement | Composite plies arrangement | Type of composite adhesive joint |
|-----------------------------|-----------------------------|---------------------------------|
| $0^\circ/0^\circ/45^\circ$  | $0^\circ/0^\circ/0^\circ$   | III-I                            |
| $0^\circ/0^\circ/45^\circ$  | $0^\circ/45^\circ/0^\circ$  | III-II                           |
| $0^\circ/0^\circ/45^\circ$  | $45^\circ/0^\circ/0^\circ$  | III-III                          |

**Table 6. Adhesive joints – Group 4.**

| Composite plies arrangement | Composite plies arrangement | Type of composite adhesive joint |
|-----------------------------|-----------------------------|---------------------------------|
| $0^\circ/0^\circ/0^\circ$   | $0^\circ/0^\circ/0^\circ$   | I-I                              |
| $0^\circ/45^\circ/0^\circ$  | $0^\circ/45^\circ/0^\circ$  | II-II                            |
| $0^\circ/0^\circ/45^\circ$  | $45^\circ/0^\circ/0^\circ$  | III-III                          |
3. Experimental results

3.1. Results
Tables 7 – 10 present the shear strength of adhesive joints and statistical analysis results of the samples from four groups of adhesive joints. The designation in tables 7-10 is the same like in the legend which was presented in table 7.

| Adhesive joints | \( S^a \), MPa | \( F^b \) | \( F^c \) | Result | \( T^{d \alpha} \) | \( T^e \) | Conclusion |
|-----------------|---------|--------|--------|--------|--------|--------|-----------|
| I-I/I-II        | 4.66    | 9.28   | 3.33   | \( \sigma_{11}^2 = \sigma_{12}^2 \) | 1.943  | -1.585 | \( \mu_{11}^2 = \mu_{12}^2 \) |
| I-II/I-III      | 4.99    | 9.28   | 1.68   | \( \sigma_{12}^2 = \sigma_{13}^2 \) | 1.943  | 1.710  | \( \mu_{12}^2 = \mu_{13}^2 \) |
| I-I/I-III       | 4.71    | 9.28   | 2.00   | \( \sigma_{11}^2 = \sigma_{13}^2 \) | 1.943  | -0.223 | \( \mu_{11}^2 = \mu_{13}^2 \) |

\( ^a \) Value of shear strength of adhesive joints.
\( ^b \) Value of statistic – \( F^b \) (Fisher Snedecor test).
\( ^c \) Obtained value of statistic – \( F^c \).
\( ^d \) Value of statistic – \( T^{d \alpha} \) (t-Student test).
\( ^e \) Obtained value of statistic – \( T^e \).
\( \sigma^2 \) – variance.
\( \mu \) – mean value.

| Adhesive joints | \( S_n \), MPa | \( F^a \alpha \) | \( F \) | Result | \( T^\alpha \) | \( T \) | Conclusion |
|-----------------|--------|--------|--------|--------|--------|--------|-----------|
| I-II/I-III      | 4.66   | 6.59   | 1.25   | \( \sigma_{12}^2 = \sigma_{23}^2 \) | 1.895  | -0.558 | \( \mu_{12}^2 = \mu_{23}^2 \) |

\( ^a \) Value of statistic – \( C^a \alpha \) (Cochran-Cox test).
\( ^c \) Obtained value of statistic – \( C \).

The statistical analysis results of the samples from Group 1 (table 3) demonstrate that the hypothesis about equality of variances of two populations is met in all cases. The hypothesis about equality of means is also confirmed in all cases, so the means in these cases are statistically equal (4.66 = 4.99, 4.99 = 4.71, 4.66 = 4.71) – table 7.

The statistical analysis results of the samples from Group 2 (table 4) demonstrate that the hypothesis about equality of variances of two populations is met in one case (I-II and II-III), while it is rejected in the remaining two cases (I-II and II-I, II-II and II-III). The hypothesis about equality of means is also confirmed in all cases, so the means in these cases are statistically equal (4.99 = 4.83, 4.99 = 5.07, 4.83 = 5.07) – table 8.

The statistical analysis results of the samples from Group 3 (table 5) demonstrate that the hypothesis about equality of variances of two populations is met in all cases. However, the hypothesis about equality of means is confirmed only in one case (I-III and III-III), so the mean in this case is statistically equal (4.71 = 4.43); however, in the remaining cases (I-III and II-III, II-III and III-III), the hypothesis is rejected, so the means in these cases are statistically different (4.71 ≠ 5.0, 5.07 ≠ 4.43) – table 9.
Table 9. Statistical analysis results of samples in Group 3.

| Adhesive joints | S, MPa | Fα  | F       | Result  | Tα   | T      | Conclusion         |
|-----------------|--------|------|---------|---------|------|--------|-------------------|
| I/II/III        | 4.71   | 6.95 | 1.33    | σ11^2 = σ22^2 | 1.895 | -2.245 | μ11^2 ≠ μ22^2      |
| II/III/III      | 5.07   | 6.39 | 1.50    | σ22^2 = σ33^2 | 1.859 | 4.040  | μ22^2 ≠ μ33^2      |
| I/III/III       | 4.43   | 6.59 | 1.13    | σ13^2 = σ33^2 | 1.895 | 1.561  | μ13^2 = μ33^2      |

Table 10. Statistical analysis results of samples in Group 4.

| Adhesive joints | S, MPa | Fα  | F       | Result  | Tα   | T      | Conclusion         |
|-----------------|--------|------|---------|---------|------|--------|-------------------|
| I/I/II          | 4.66   | 6.59 | 2.63    | σ11^2 = σ22^2 | 1.859 | -0.499 | μ11^2 = μ22^2      |
| II/I/III/III    | 4.83   | 6.39 | 2.67    | σ22^2 = σ33^2 | 1.859 | 1.371  | μ22^2 = μ33^2      |
| III/I/I         | 4.43   | 6.59 | 1.78    | σ33^2 = σ11^2 | 1.895 | 1.084  | μ33^2 = μ11^2      |

The statistical analysis results of the samples from Group 4 (table 6) demonstrate that the hypothesis about equality of variances of two populations is met in all cases. The hypothesis about equality of means is confirmed in all cases, too, so the means in these cases are statistically equal (4.66 = 4.83, 4.83 = 4.43, 4.66 = 4.43) – table 10.

3.2. Discussion

The results of adhesive joint strength demonstrate that the arrangement of composite plies affects the strength of the investigated adhesive joints. In addition, it can be observed that in the adhesive bonding of composite materials the strength of adhesive joints is also significantly affected by the number of fiberglass fabric plies.

It must be stressed that special care was taken to ensure that all composite materials are made under the same conditions, the only variables being the arrangement and number of plies. Based on the statistical analysis of the strength test results, it is found that in most cases there are no statistically significant differences in the yielded means of strength (Groups 1, 2, 4). However, the results for other tested samples (Groups 3,) reveal that such differences do occur. Group 3 involved analysis of the effect of shifting the turned ply on the one side of the adhesive layer, while the plies arrangement on other side was unchanged and had the form of 0°/0°/45° (the turned ply was located closest to the adhesive layer, table 5). Supported by a statistical analysis, these results demonstrate a clear effect of the fibreglass fabric plies arrangement on failure force. The best results in this group are yielded for the configuration II-III (plies arrangement: 0°/45°/0°/adhesive/45°/0°/0°). On the other hand, the worst results are obtained for the configuration III-II, where the plies arrangement is changed only at the adhesive layer (plies arrangement: 0°/0°/45°/adhesive/45°/0°/0°). The results in Group 4 demonstrate that the symmetrical plies arrangement has no significant effect on the strength of the examined adhesive joints. Nonetheless, it is observed that changing the ply arrangement in the vicinity of the adhesive layer (joint III-II) has a negative effect on the strength of this layer, as opposed to the case when the ply arrangement is not changed (joint I-I). However, the combination of these two plies arrangement types into one (joint I-II) reveals a certain synergy and yields better results than in the case of the two previous adhesive joints. Summing up, plies arrangement in composite materials affects the strength of adhesive joints made of these composites.
4. Summary

The paper investigated the effect of plies arrangement in a hand-made composite material on the strength of adhesive joints made from this composite. The significance of this problem is particularly true these days owing to a wide use of composite materials to design structures with required specific strength. To investigate the effect of plies arrangement on the strength of fibreglass fabric, 3 composite materials were fabricated, each having a different plies arrangement. Special care was taken to maintain the same manufacturing conditions throughout the entire process as well as to ensure that the composite material be produced with utmost accuracy so as to yield the most reliable results. Individual composite plates were cut into smaller samples and then adhesive bonded using an epoxy adhesive made from Epidian 61 epoxy resin combined with triethylenetetraamine hardener. The stabilizing period of all samples was set to 7 days. After that, each sample was measured and subjected to failure tests. Some of the procedures had to be repeated because of a significant scatter of the results. The final results were then divided into groups to examine the effect of different variants of plies arrangement. Summing up, it was found that in some cases (joint II-III) plies arrangement has a significant effect on increasing the shear strength of samples. Two types of adhesive joints samples exhibit higher shear strength: those consisting of two samples of adhesive joints made of uniform thickness composites as well as those with alternate plies arrangements. It should be noted that the both tangential plies in the plies arrangement 0°/adhesive layer/0° should not be turned in the vicinity of the adhesive layer. But Composite II (0°/45°/0°) samples exhibit the lowest tensile strength. In this composite central ply has the 45° orientation. However, the combination of two plies arrangement types into one (joint II-II) reveals a certain synergy and yields better results than in the case of the joint III-III and joint I-I. Concluding, plies arrangement is of vital importance and should be taken into account when designing composite materials for adhesive bonding. Nonetheless, this problem has not been exhaustively investigated in this paper. It can be studied further using samples which differ from those used in this study (e.g. samples with 45°/0°/45° plies arrangement).

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