Comparative Analysis of Mechanical Properties of Laminated Carbon Fiber Reinforced Composites Based on PEEK and PI Matrices

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Abstract. The mechanical properties of laminated carbon fiber reinforced composites based on PEEK and PI matrices have been studied. Various schemes of carbon fiber lay-outing were employed. Flexural stress-strain curves, optical images of fracture surfaces and SEM micrographs of composite structure were analyzed. It has been shown that laminated composites reinforced with continuous unidirectional fibers according to layout scheme \([0^\circ/0^\circ]\) possessed the maximum values of mechanical properties (flexural modulus and bending strength) under three-point bending. PEEK based composites exhibited 2 times higher flexural strength in contrast to PI based ones (0.4 GPa and 0.2 GPa, respectively), while the modulus of elasticity in bending was 4 times larger (61 GPa and 15 GPa, respectively).

1. Introduction
Research and development of polymer-based structural composites are critical trends in advanced industries worldwide. This topic is most relevant for the aerospace industry, which requires high specific strength values (the ratio of material strength to density) of the materials used. Currently, R&D is being actively carried out on the reinforcement of thermoplastic polymers reinforced with continuous fibers, which makes it possible to substantially increase mechanical properties of the composites [1-2].

The loading of polymer matrix with short fibers provides an increase in strength properties compared to neat polymer, as well as the possibility of processing by technologies traditional for most thermoplastics, i.e. extrusion and injection molding. However, there is a noticeable decrease in plasticity and corresponding embrittlement of the material, which severely limits the use of composites with short fibers. It was shown in [4] that modulus of elasticity of polymer composites increases sharply with enlargement of the length of carbon fibers.

Loading with chopped fibers (from a millimeter to tens of millimeters length) is an effective way to improve tribological and mechanical properties of composites based on polyetheretherketone (PEEK) [5-7]. The reinforcement with continuous fibers can significantly improve the performance of composites based on thermoplastic matrices. Among industrially employed continuous fibers are carbon, glass, basalt, metal ones. They have the shape of ribbons, tows or fabric, etc. [8]. The technology for producing parts of
thermoplastic composites reinforced with continuous fibers consists in layering a binder (in the form of a polymer powder or a film) and fibers. For example, in [9] the properties of a layered composite consisting of 9 layers of a PEEK film (1000-300G) with a thickness of 300 μm (produced by the Victrex) and 8 layers of 3K-T300-plain carbon fiber (produced by the Toray Industries) were studied. The stacking scheme was [0°/90°]. The fabricated composite had the following properties: ultimate bending strength of 400 MPa, flexural modulus of 20 GPa, and interlayer shear strength of 50 MPa. The highest mechanical properties of polyimide (PI) based composites were also achieved through a continuous fiber reinforcement.

The different arrangement of fibers gives rise to characteristic changes in mechanical properties, as well as to failure pattern of composite materials [10]. Mechanical properties of carbon composites with various modes of weaving and laying were studied in [11]. It has been shown that mechanical properties of the cross-reinforced composite [0°/90°/45°/-45°] are 30-40% lower compared to unidirectional composite [0°/0°]. However, authors of paper [12] noted that the anisotropy of fiberglass composites was evident when the weaving and stacking mode was [0°/0°], while the composite [0°/90°/45°/-45°] possessed isotropic properties.

The aim of this paper was to study the mechanical properties (in particular flexural modulus and bending strength) of laminated polymer composites based on polyetheretherketone and polyimide matrix, reinforced with continuous carbon fibers at variation of layout schemes.

2. Materials and methods

PEEK powder (Victrex, PEEK 450 PF, UK) with an average particle size of 50 μm and PI powder (Solver, PI-Powder, China) with an average particle size 16 μm were employed. Unidirectional carbon fabric (tape FibArm 12K-300-230, Umatec, Russia) with areal density 230 g/m² and tensile strength above 4.9 GPa as well as biaxial carbon fabric (0/90-50K-1270-106, HC "Composite", Russia) with surface density 106 g/m² were used as a reinforcing filler.

The ratio of the components of the fabricated polymer composites (40 vol. % fiber / 60 vol. % matrix) was selected according to literature data, as the most effective for increasing mechanical characteristics.

Various layout schemes were employed (Figure 1).

![Figure 1. Layout schemes for carbon fiber layers (polymer powder layers are located between them).](image)

After placing the layered components in the mold, the composite samples were prepared by hot pressing with the use of GT-7014-A hydraulic press (GOTECH Testing Machines Inc., Taiwan) at specific pressure of 10 MPa and a temperature of 400 °C. Cooling rate was 2 °C/min.

Shore D hardness was measured with the help of Instron 902 series machine (Instron, USA) in accordance with ASTM D2240. Mechanical properties of the composites were determined under three-point
bending on an electromechanical testing machine Instron 5582 series (Instron, USA) in accordance with ASTM D790-10. Stripe shapes samples had the size of 70×10×3.5 mm. Optical microscope Neophot 2 (Carl Zeiss, Germany) was used to examine the cross-section of fractured samples (after 3-point bending). Investigations of polymer composite structures were carried out in accordance with STO TSU 041-2009 “Technique for studying the structure of solid surface by scanning electron microscopy” using a system with electron (focused) beams Quanta 200 3D (FEI Company, USA); accelerating voltage was 20 kV. Copper film was preliminary deposited onto fracture surface of composite samples.

3. Results and discussion

Figure 2 shows the diagrams of flexural stress versus relative flexural strain. Neat PEEK and PI had comparable mechanical properties (curves 1, 6). However, they decreased sharply when loaded with fibers (curves 3, 8) as well as at cross-stacking of reinforcing layers (4, 9). Biaxial reinforced composites (curves 5, 10) exhibited mechanical properties 15-20% lower compared to unidirectional reinforcement. Composites with unidirectional reinforcement had the highest mechanical properties and largest deformation when loaded transverse to the fiber orientation. The composite PEEK/C [0°/0°] (2) had a transverse flexural modulus of 61.7 GPa, while in composites PI/C [0°/0°] (7) it was equal to 15.6 GPa.

![Figure 2](image_url)

**Figure 2.** Flexural stress-strain curves of PEEK (1); PEEK/C [0°/0°] (2); PEEK/C [90°/90°] (3); PEEK/C [0°/90°] (4); PEEK/C [biaxial] (5); PI (6); PI/C [0°/0°] (7); PI/C [90°/90°] (8); PI/C [0°/90°] (9); PI/C [biaxial] (10).

Detailed data on physical and mechanical properties of all tested samples are presented in Table 1. It is seen that the largest increase in mechanical properties (flexural modulus and bending strength at three-point bending) was observed in composite samples # 2 and # 7, reinforced according to the [0°/0°] scheme.

Optical images of fractured sample cross section tested at 3-point bending are shown in Figure 3. Optical images of neat PEEK and PI samples are not presented since no cracking happened there.
Table 1. Physical-mechanical properties of carbon reinforced composites and unreinforced polymers.

| Sample       | Density, g/cm³ | Shore D hardness | Flexural modulus $G_H^f$, GPa | Bending strength $\sigma_H^f$, MPa | Strain $\varepsilon_{max}$ |
|--------------|----------------|-----------------|-------------------------------|----------------------------------|---------------------------|
| PEEK         | 1.30           | 80.1±1.7        | 4.2±0.6                       | 166.2±6.1                       | 0.050±0.005               |
| PEEK/C [0°/0°] | 1.54           | 81.5±0.6        | 61.7±1.0                      | 422.8±12.5                      | 0.050±0.001               |
| PEEK/C [90°/90°] | 1.51           | 81.4±0.6        | 4.3±0.2                       | 42.3±6.4                        | 0.015±0.004               |
| PEEK/C [0°/90°] | 1.46           | 80.4±0.8        | 21.0±3.4                      | 173.8±26.8                      | 0.022±0.001               |
| PEEK/C [biaxial] | 1.33           | 82.0±0.7        | 41.2±1.8                      | 277.2±34.9                      | 0.050±0.001               |
| PI           | 1.37           | 80.3±0.5        | 4.01±0.5                      | 143.3±6.2                       | 0.050±0.001               |
| PI/C [0°/0°]  | 1.40           | 76.8±1.5        | 15.6±0.9                      | 190.2±18.7                      | 0.050±0.001               |
| PI/C [90°/90°] | 1.43           | 82.4±1.3        | 4.2±0.3                       | 30.1±4.1                        | 0.023±0.001               |
| PI/C [0°/90°]  | 1.33           | 70.2±2.9        | 4.3±0.5                       | 43.0±5.2                        | 0.050±0.001               |
| PI/C [biaxial] | 1.38           | 78.6±1.2        | 11.9±1.3                      | 158.4±1.2                       | 0.040±0.001               |

Figure 3. Optical images of cross section view of 3-point bending fractured samples:
PEEK/C [0°/0°] (a); PEEK/C [90°/90°] (b); PEEK/C [0°/90°] (c); PEEK/C [biaxial] (d);
PI/C [0°/0°] (e); PI/C [90°/90°] (g); PI/C [0°/90°] (g); PI/C [biaxial] (h).

It was found that cracking was slightly exhibited when sample fabrication followed [0°/0°] layout (figure 3, a, b). Longitudinal delamination occurred, while interlayer cracks took ~1/5 of the sample thickness. Through transverse cracking occurred when lay-outing out was [90°/90°] (figure 3, b, f). Tensile, shear and delaminating components of cracking were evident. Pronounced interlayer delamination took place when the [0°/90°] scheme (c, g) was employed. Exhibited shear cracking were characteristic when the [biaxial] scheme (d, h) was employed.
SEM micrographs of composite structures are shown in Figure 4. One can see that regions with incomplete wetting of carbon fibers presented in the composites. Thus, in order to further increase strength properties the problem of matrix material better penetration into fiber layers should be solved.

**Figure 4.** SEM micrographs of composite structures: PEEK (a), PEEK/C [0°/0°] (b); PEEK/C [90°/90°] (c); PEEK/C [0°/90°] (d); PEEK/C [biaxial] (e); PI (f); PI/C [0°/0°] (g); PI/C [90°/90°] (h); PI/C [0°/90°] (i); PI/C [biaxial] (j).

4. Conclusions

It has been shown that laminated composites reinforced with continuous unidirectional fibers according to layout scheme [0°/0°] possessed the maximum mechanical properties (flexural modulus and bending strength) under three-point bending. PEEK based composites possessed flexural strength 2 times higher compared to PI based ones (0.4 GPa and 0.2 GPa, respectively), while the modulus of elasticity in bending is 4 times larger (61 GPa and 15 GPa, respectively).

According to the obtained results the use of PI powder for fabricating laminated carbon fiber reinforced composites is of low prospect because of low mechanical properties (in comparison with PEEK based composites). The most probable reason is the lower melt flow index of the PI matrix.

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