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# THE MECHANICAL PROPERTIES IMPROVEMENT OF THERMOPLASTICS-BASED FIBER METAL LAMINATES

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**Abstract.** This study investigated the possibility of increasing the fiber metal laminates mechanical properties by improving the impregnation quality due to the addition of a different number of layers of polymer film on the prepreg-metal interface. Specimens were made based on two thermoplastic polymers (polyamide-6 and polypropylene), glass fibers (E-glass), and aluminum alloy (AlMg6) by hot pressing. The theoretical calculation was performed for the tensile strength according to the rule of mixtures. The dependences of tensile strength, impact strength, and pattern failure modes on the fiber metal laminates structure and polymer type were produced. Options for improving metal-polymer adhesion up to 2.7 times by different methods of aluminum surface modification were shown.
**Keywords:** thermoplastic polymers, fiber metal laminate, rule of mixtures, prepreg, composite

## 1. Introduction

During the last decades, composite materials are the subject of constant interest for researchers all over the world. The developments in this field allowed to achieve a significant weight reduction of the structures while maintaining their high mechanical characteristics, as well as to provide high fatigue properties and corrosion resistance. All these advantages have contributed to the expansion of composite materials application in the aerospace industry [1-3].

In the late 1970s at the Technical University of Delft (Netherlands) it was shown that, instead of using bulk monolithic materials, laminates consisting of thin layers of dissimilar materials (Fiber Metal Laminate – FML), it is possible to achieve a significant reduction in the crack growth rate. With the crack nucleation in one of the layers at the interface, its growth slows down and this effect continues until the crack appears in the next layer [4]. Based on these studies, in 1978 the first layered metal-polymer composite material system called ARALL (Aramid Reinforced Aluminum Laminate) was developed – based on aramid fibers impregnated with epoxy resin and aluminum alloy 2024-T3 with a thickness of 0.3 mm [3].

In subsequent years, three main types of FML composites were developed: ARALL generation 1-4 (based on aramid fibers), CARAL (based on carbon fibers), and GLARE generation 1-6 (based on glass fibers), which were made based on various aluminum alloys. One of the features of these materials is that they are all made with epoxy binders, which have low fracture toughness, and this is especially critical for FML composites [5].

Thermoplastic polymeric materials are of great interest for aerospace and automotive applications due to their properties combination such as high fracture toughness, high

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mechanical strength, resistance to the external environment, and recyclability. All these advantages allow them to gradually displace thermoset binders from the market [6,7].

At present, for the production of prepregs based on thermoplastic polymers, the following methods are commonly used: dry powder or powder suspension impregnations with subsequent sintering, melt impregnation, and fiber commingling to produce hybrid yarns [7-8]. In this case, in the process of prepreg production, a polymer is often applied to the surface of the fabric or unidirectional fibers, and the final impregnation is carried out during the product molding during hot pressing or vacuum consolidation. For example, Patou et al. investigated the effects of vacuum consolidation modes on the porosity and mechanical properties of CF-PPS composites. As a result, they showed that, depending on the technological parameters, the porosity could vary from 1 to 13 %, which leads to a decrease in interlaminar shear strength up to 35% [9].

This study was aimed to investigate the improving possibility of the impregnation and the formation qualities, to obtain the thermoplastic prepreg with higher mechanical properties during the FML production by hot pressing. For this purpose, the influence of the prepreg layers stacking as well as polymer nature and amount, on the mechanical properties of the resulting material was studied.

**Experimental procedure.** To produce composite materials were used aluminum sheets, 0.5 mm thick of AlMg6 alloy; 2 layers of prepreg, with $0^o/90^o$ layup, based on glass fiber and two types of polymer, polyamide (GPA6H-60-430-std) and polypropylene (GPP65U-250-std), manufactured by Qiyi Technology, China; Polymer films: polyamide-6 (PA) – 50 µm thick and polypropylene (PP) – 40 µm thick (Table 1).

Table 1. Prepregs components properties

| Material | Tensile strength | Density | Width |
|---|---|---|---|
| GPA6H-60-430-std | >800 | 1.71 | 0.17 |
| GPP65U-250-std | >800 | 1.58 | 0.25 |
| AlMg6 | 315 | 2.64 | 0.5 |
| PA film | 65 | 1.13 | 0.05 |
| PP film | 40 | 0.91 | 0.04 |

**Aluminum surface treatment.** To study the effect of the aluminum surface treatment, 3 methods were studied: etching in 32% $HNO_3$ solution; P2 treatment – etching in $Fe_2(SO_4)_3$ and $H_2SO_4$ solution; sulfuric acid anodizing (SAA). Treatment modes are presented in Table 2.

Before surface treatment, the aluminum sheets were pretreated by washing in a detergent; rinsing in acetone; etching in a 10% NaOH solution for 10 seconds. After each step, the aluminum was washed with distilled water.

Table 2. Metal surface treatment methods

<table>
<tr><th>#</th><th>Name</th><th>Treatment</th><th>Solution</th><th>Temp.</th><th>Time</th><th>Details</th></tr>
<tr><td>1</td><td>$HNO_3$</td><td rowspan="2">Chemical</td><td>32% $HNO_3$</td><td>20°C</td><td>1 min</td><td>-</td></tr>
<tr><td>2</td><td>P2</td><td>$Fe_2(SO_4)_3$ 127 g/l<br>$H_2SO_4$ 185 ml /l</td><td>65°C</td><td>8 min</td><td>-</td></tr>
<tr><td>3</td><td>SAA</td><td>Electro-chemical</td><td>$Al_2(SO_4)_3$ 200 g/l<br>$H_2SO_4$ 60 ml/l</td><td>20°C</td><td>20 min</td><td>Current density 1.5 $A/dm^2$</td></tr>
</table>

**FML specimens fabrication**. Composite materials were made by hot pressing at a pressure of 2 MPa and temperature 225°C for PA and 190°C for PP, holding time at the pressing temperature was 20 min. Two layers of prepreg with 0° and 90° orientation were placed between the sheets of aluminum. The influence of the polymer film number on the interface between aluminum and prepreg (from 0 to 3 layers) and the additional layer between two prepregs were also investigated. To study the influence of the layer structure on the properties of CM specimens with different prepreg and film combinations were made: GF-PA – PA film; GF-PP – PA film and GF-PP – PP film (Fig. 1). The produced CM specimens had a thickness from 3 to 5 mm.

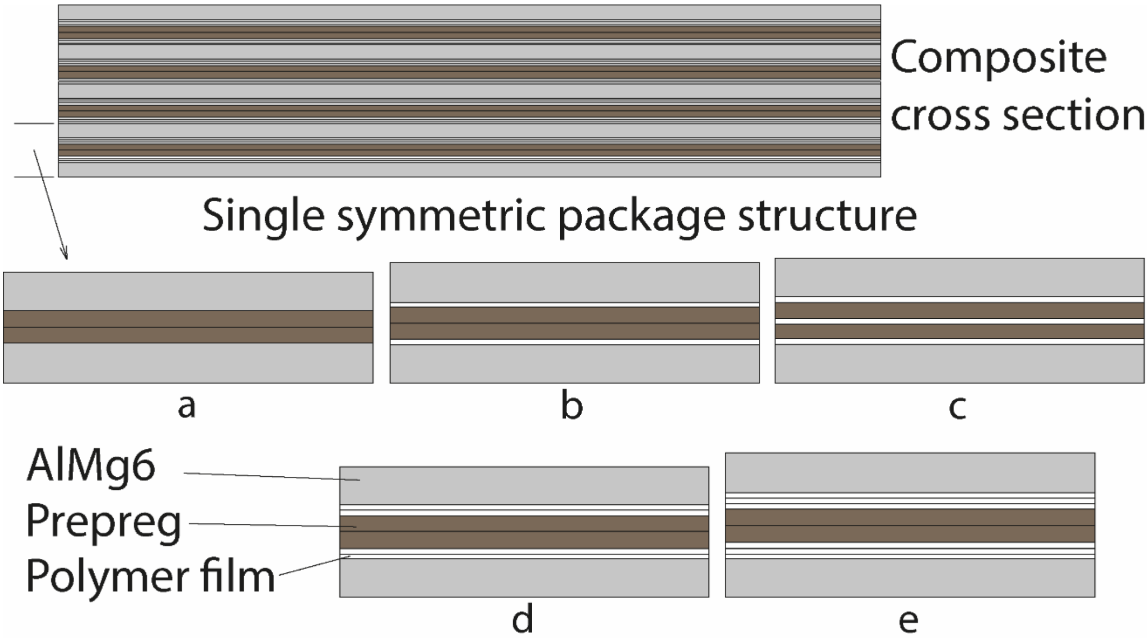


**Fig. 1.** The layers structure according to layers number. a – 0, b – 2, c – 3, d – 4, e – 6

**Testing methods.** Tensile testing was performed on a ZwickRoell Z100 universal testing machine, strain rate was 2 mm/min, the strain was determined using contact extensometers. Impact strength testing was performed according to ISO 179-1, on a ZwickRoell RKP 450, with hammer energy of 150 Joules. Adhesion was tested by measuring the tensile shear strength of the aluminum-polymer bonding contact according to ASTM D 1002

## 2. Results and discussions

**Tensile strength.** Figure 2 shows the most representative experimental curves of the specimens in the tensile test. The modulus is almost independent of polymer type and configuration, having a value of 50±3 GPa for all specimens.

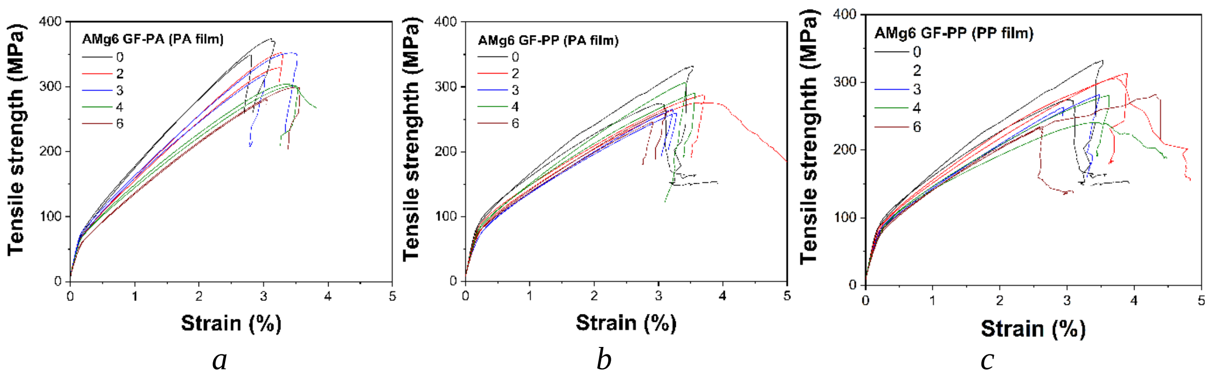


**Fig. 2.** Tensile stress-strain curves. a – PA-PA; b – PP-PA; c – PP-PP

The theoretical strength of FML was calculated by the rule of mixtures [10]:

$$\sigma_{FML} = \sum_{i=1}^{n} \sigma_i \frac{S_i}{S_{FML}},$$

where, $\sigma_{FML}$, $\sigma_i$ – strength of FML and component respectively, $n$ – number of components, $S_{FML}$, $S_i$ – cross-sectional area of FML and component respectively.

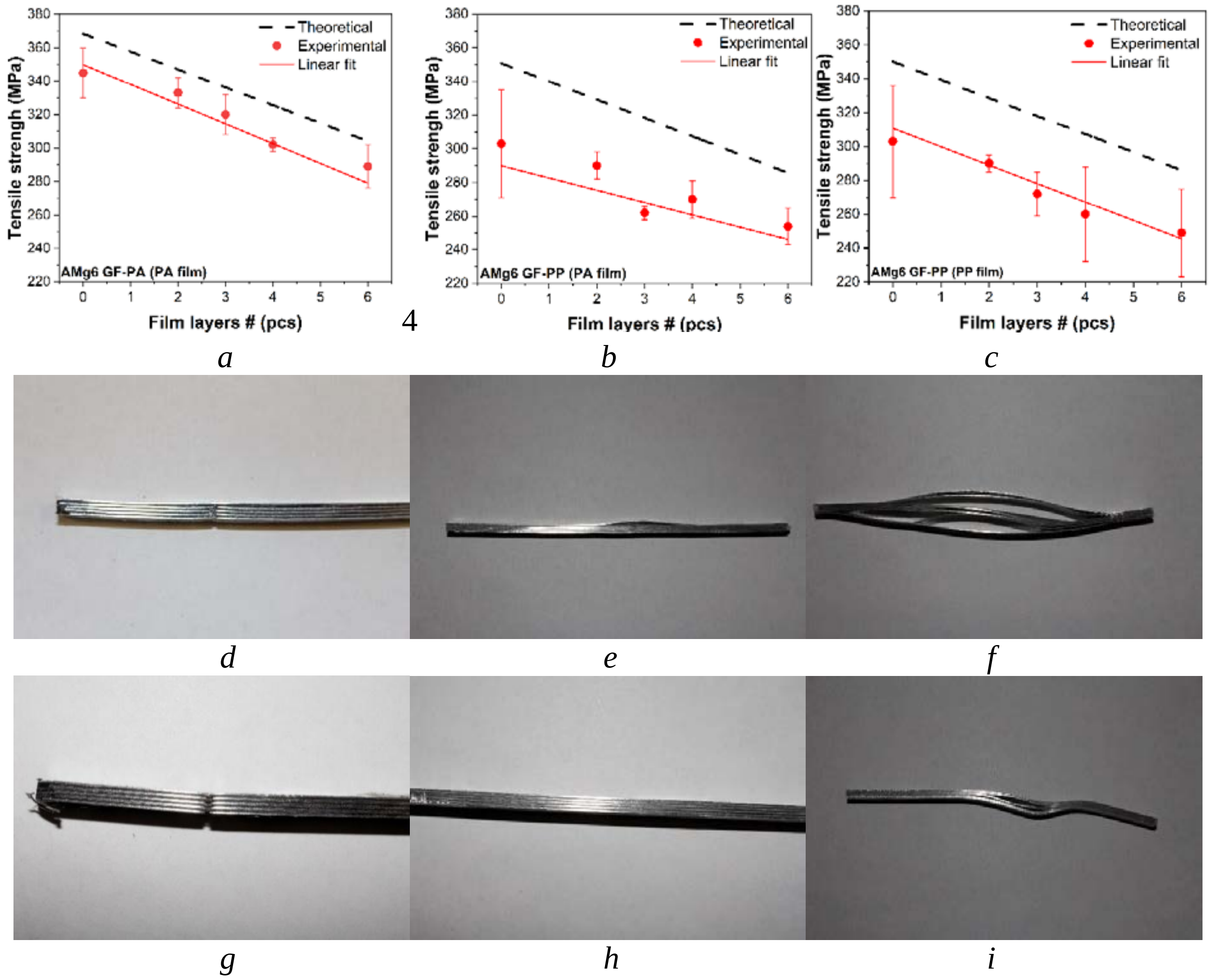


**Fig. 3.** Tensile strength vs. a number of layers of polymer film and pictures of specimens after the test. a, d, g – PA-PA film; b, e, h – PP-PA film; c, f, i – PP-PP film

Figures 3(a-c) show a comparison of the experimental and theoretical strength dependences of FML specimens, depending on the film layers number. As can be seen, the slopes of the experimental data coincide quite exactly with the theoretical ones based on the rule of mixtures. The photos of the specimens after the test (Fig. 3(d-i)) clearly show that depending on the polymer, the specimen fracture mode changes greatly.

For PA-PA specimens, the similarity between the theoretical calculations and the real data is rather high (deviation of the order of 5%). This high agreement is presumably due to the high adhesion between the layers. As can be seen in Fig. 3(d), (g) the specimens did not delaminate after the test.

The PP-PA specimens demonstrate a loss of performance for 3 film layers, which can be attributed to the deterioration of the adhesive contact between prepregs. In this case, the deviation already reaches 13%. We assume that in this case there is destruction by defects at the PP-PA interface because the polymers do not interact well with each other.

In the PP-PP plot, the deviation reaches 19%. This is presumably due to poor adhesion between Al and PP since PP is an inert polymer. It is clearly seen from the photographs where deformation of the specimen leads to delamination of the laminate at the metal-polymer interface, due to which the specimen does not reach the theoretical strength.

**Impact strength.** Figure 4 shows the dependence of material impact strength on the polymer film layers for different combinations of prepreg and film, as well as photos of specimens after the test.

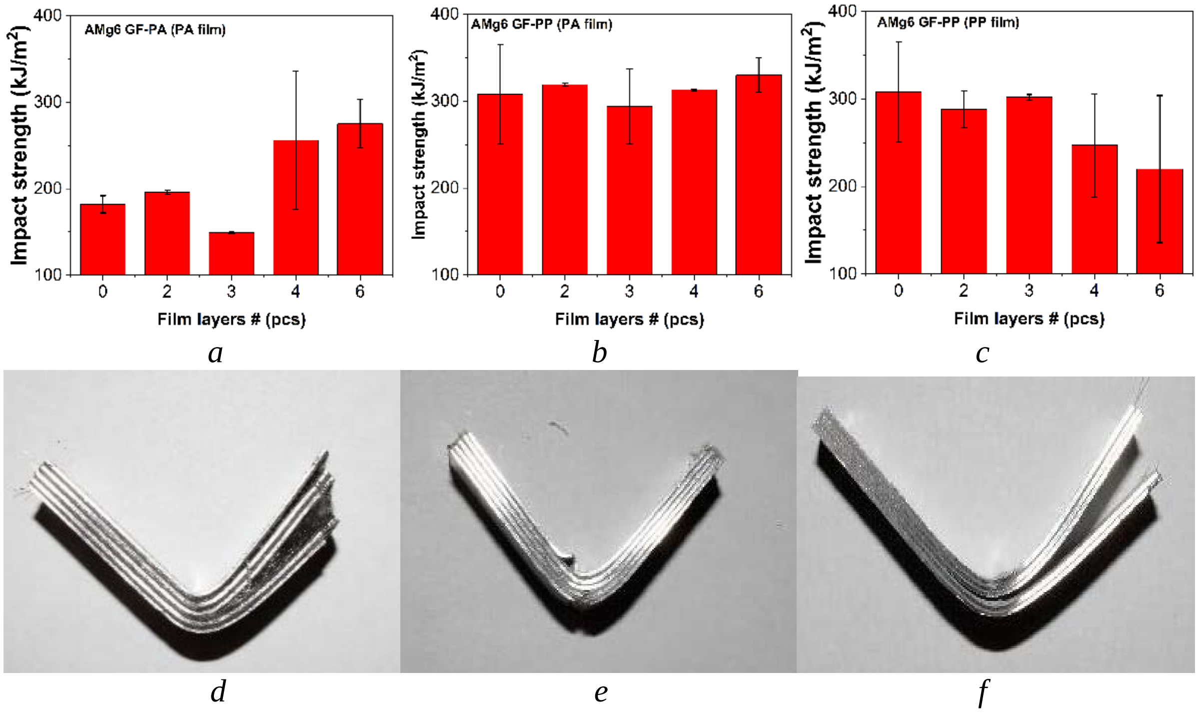


**Fig. 4.** Impact test results of CM specimens, a, d – PA-PA film; b, e – PP-PA film; c, f – PP-PP film

The PA-PA specimens in this test show the worst result among others. When analyzing the failure mechanism of the PA-PA specimen (Fig. 4(d)), it is clear that the failure occurs through a multiple shear mechanism. It is worth noting that ply shear occurs in almost every ply. It is also clearly seen that the fracture occurred not at the metal-polymer boundary but inside the prepreg layer. The observed fracture mechanism may be related to the fact that after pressing the specimens, a large number of pores were formed in the prepreg layer due to the high viscosity of the PA6 melt, which leads to low interlayer strength.

An increase in the number of film layers increases the impact strength value. For specimens in which the film was added at the metal-polymer interface, the impact strength increases linearly with the number of layers. This is because a significant increase in polymer concentration leads to an increase in the quality of the prepreg impregnation during pressing.

The fracture pattern of PP-PA specimens is normal for this class of material. Compressive stresses, at the point of impact, result in buckling in the aluminum, since it is relatively ductile. Tensile stresses, on the opposite side, cause the layers of both aluminum and prepreg to rupture. The impact strength of FML with prepreg based on PP is significantly higher than that of PA prepreg since polypropylene has a much lower melt viscosity compared to polyamide, which results in a composite structure with less porosity and fewer defects.

Some PP-PP specimens were fractured by the shear mechanism at the metal-polymer interface, some by the mechanism described for PP-PA specimens. This is due to the low value of adhesion strength of polypropylene to metal. In general, the value of impact strength for specimens with PP prepreg is at the level of 300 kJ/m$^2$ and is weakly dependent on the number of films. Specimens with 4 and 6 layers of polypropylene film, at first glance, show a decrease, but they have a large deviation due to delamination.

**Adhesion metal-polymer.** To improve the composite properties, the possibility of increasing the strength by increasing the adhesion between the polymer and aluminum was investigated. For this purpose, the aluminum was processed with 3 methods of surface treatment

Since the polyamide prepreg showed a high value of experimental strength relative to the theoretical but failed on the polymer layer under impact test, and the mixture of polyamide and polypropylene has additional boundaries affecting the test results, the adhesion study was carried out for PP-PP specimens. The result is shown in Fig. 5.

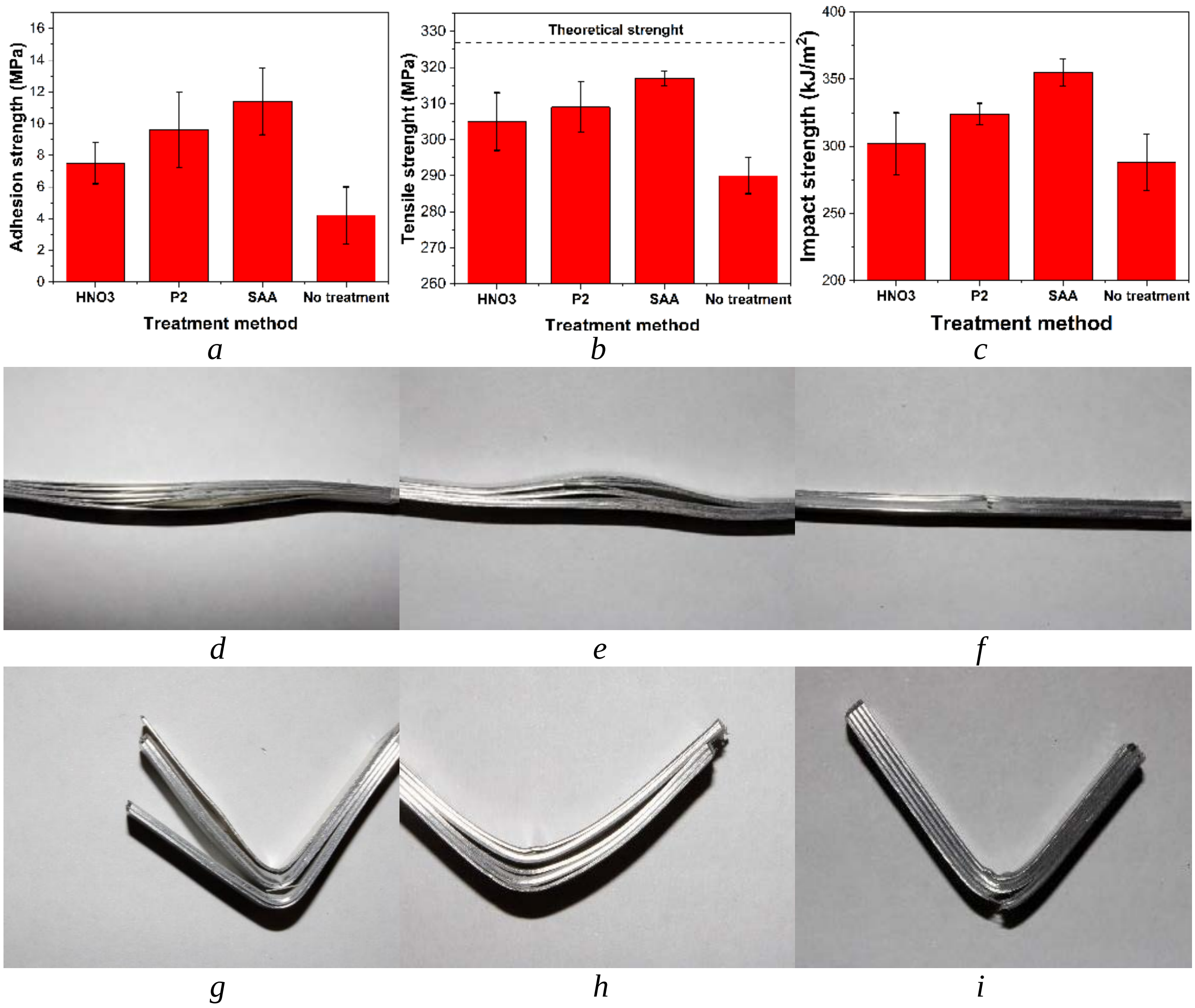


**Fig. 5.** Dependence of shear strength (a), tensile strength (b), and impact strength (c) on the type of surface treatment. Specimen photos after tensile and impact tests, respectively: d, g – $HNO_3$; e, h – P2; f, i – SAA

Figure 5a shows that all surface treatments can increase the adhesion strength at the polymer-metal interface. Among the chemical surface treatments, the P2 method can increase the adhesion strength by more than 2 times compared to the untreated surface. SAA allows raising the adhesion strength by 2.7 times.

The specimen photos after the test show delamination with less intensity, and it depends on the type of treatment. These results are in good agreement with the results of the adhesion strength test (Fig. 5(a)). Figures 5(d-f) show that with the increase of the adhesion strength, the delamination after tensile decreases, due to better load redistribution. The impact mechanism changes from shear at the polymer-aluminum interface to a mechanism in which compression zones with bulking of the aluminum layer and tension zones with crack formation occur, typical for PA-PP specimens.

## 3. Conclusions

FML specimens based on AlMg6 alloy, fiberglass thermoplastic prepreg, and polymer films were made. As a result of the specimens examined, it was found:

1. The properties of FML can be predicted with a high degree of confidence using the mixture rule, as long as sufficient adhesion between the components is ensured. For polyamide, the error was about 5%, for polypropylene 19%.
2. The prepreg impregnation quality has little effect on the tensile test results since the contact area of the polymer with the fiber remains quite large in absolute value. More influence is caused by the adhesion between the layers. In case of its insufficient value, the prepreg slips relative to the metal, which leads to the occurrence of local stresses that exceed the strength of the individual layer and their subsequent destruction.
3. Under impact loads, the prepreg layer structure formed during the pressing process has a decisive influence on the impact strength. Uneven distribution of polymer over the fiber leads to the FML fracture by the shear mechanism in the prepreg layer, which significantly reduces the strength properties. Improving the quality of the impregnation prevents the occurrence and development of shear cracks by changing the fracture mechanism.
4. The used methods of metal surface modification allow increasing the adhesion to the polymer by more than 2.7 times. Electrochemical treatment allowed to increase the impact strength from 288 to 355 MPa. In addition, this type of treatment made it possible to produce specimens with the smallest standard deviation of test results.

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