ISSN 1995-4212, Polymer Science, Series D, 2018, Vol. 11, No. 3, pp. 263–266. © Pleiades Publishing, Ltd., 2018.
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Adhesive-Strength Evaluation via the Pull-Out Method in a Binder—Elementary-Filament System at Various Treatments of Filaments

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Received December 13, 2017

Abstract—The adhesive strength of binders with elementary filaments that have been used for the production of LUP carbon tape covered by different types of coatings is evaluated via the pull-out method. Two types of single-layer coatings are considered, one of which is based on titanium of the BT1-0 brand while the other is made of 12Cr18Ni10Ti steel, as well as a bilayer coating composed of a titanium layer coated by a stainless-steel layer. The adhesive strength is determined for thermoreactive (epoxy with amine hardener) and thermoplastic (polyamide) types of binders. The best adhesive strength (more than 30%) of binders is achieved when the LUP carbon tape is covered with stainless steel.

Keywords: carbon tape, elementary filament, titanium and stainless-steel coatings, epoxy and polyamide binders

DOI: 10.1134/S1995421218030127

INTRODUCTION

Carbon tissues and other fibrous fillers are extensively used in the aerospace industry thanks to their unique properties, including high specific characteristics [1, 2]. Prepregs have been used to produce carbon-fiber-reinforced-plastic products for a long time that provided low porosity and, consequently, high mechanical characteristics, but their cost increased to a great extent. One powerful tool for reducing net cost is substituting prepregs with direct molding, including vacuum diffusion [3, 4].

Epoxy materials are extensively applied as binders in the aerospace industry due to their outstanding technical properties that enable the curing process to be conducted over a wide range of temperatures, even at room temperature [5]. Furthermore, epoxy binders exhibit high adhesion to most substrates, such as carbon fibers [6, 7].

There have been numerous attempts made at increasing the adhesive-strength interaction between a carbon-fibrous filler and epoxy binder using various techniques that allow either the properties of filler or those of binder to be tuned in a targeted manner. For instance, the reactivity of carbon-fibrous fillers can be improved through oxidation, while that of the binder is augmented via chemical modification, intensification of curing processes [8], and methods. One relatively new method for increasing adhesive interplay and providing new functional properties for carbon-fiber-reinforced plastics is metallization of carbon tissues (tapes).

The present work is aimed at evaluating the adhesive strength of elementary filaments used for the fabrication of LUP carbon tape before and after its coating by metal at applying thermoreactive and thermoplastic binders via the pull-out test.

Pull-out methods have become popular in Russia for the development of state-of-the-art polymer composites [9, 10].

MATERIALS AND METHODS

LUP domestic unidirectional carbon tape served as samples that were covered with three different coatings, a single-layer one of BT1-0 titanium and 12Cr18Ni10T stainless steel, a bilayer coating composed of single-layer titanium, and a surface stainless-steel layer. For this purpose, a MIR-2 magnetron sputtering setup was used. To apply coatings, LUP carbon tapes (the maximum dimensions were 297 × 210 mm) were placed in a UPM-500 plasma-chemical reactor using an MIR-2 magnetron sputtering installation. Plasma-chemical processing of samples was implemented on both sides of the tape at the same pressures, discharge currents, and exposure times.

The binder was an imported compound of epoxy resin and amine hardener the properties of which were similar to those of ED-20 epoxy resin and diethylene triamine (DETA). The binder was cured under accelerated conditions: first, the temperature was increased

(a) (b)

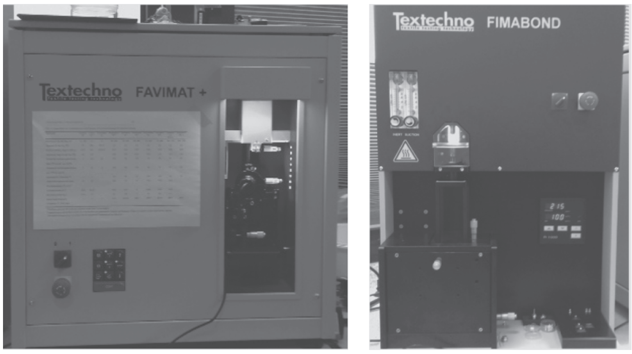

Fig. 1. Photo of (a) a pull-out testing machine with (b) a sample-preparation stand.

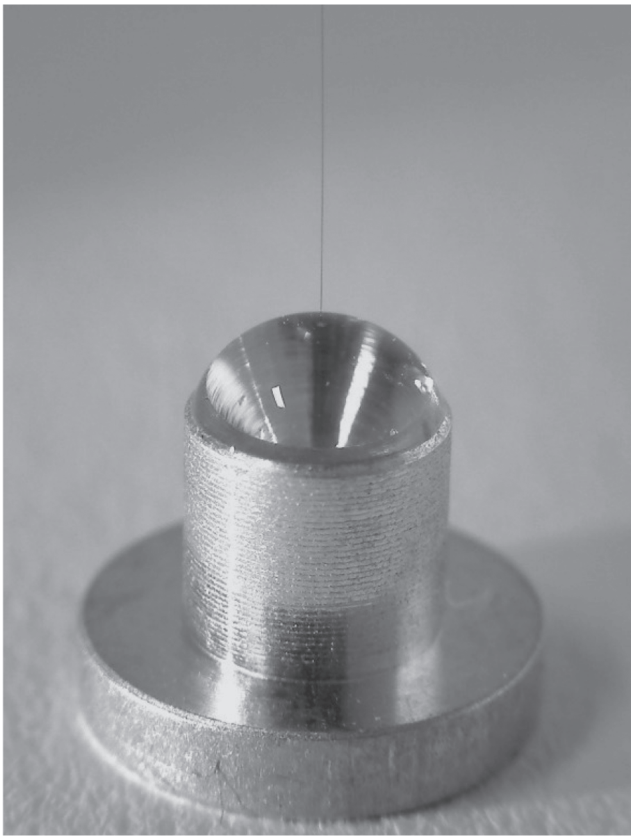

Fig. 2. Photo of a device for preparation of samples with cured epoxy binder for their subsequent pull-out tests. An elementary filament is fixed inside it.

at a rate of 25°C/min to 100°C; second, samples were exposed for 5 min; and, third, it was cooled to room temperature. The adhesive strength of a binder subjected to this curing mode is expected to much lower than in the case of curing within 24 h at room temperature. However, the determination of absolute adhesion-strength values is not within the scope of this work, although estimating their relative values at various metal coatings atop the elementary filament com-

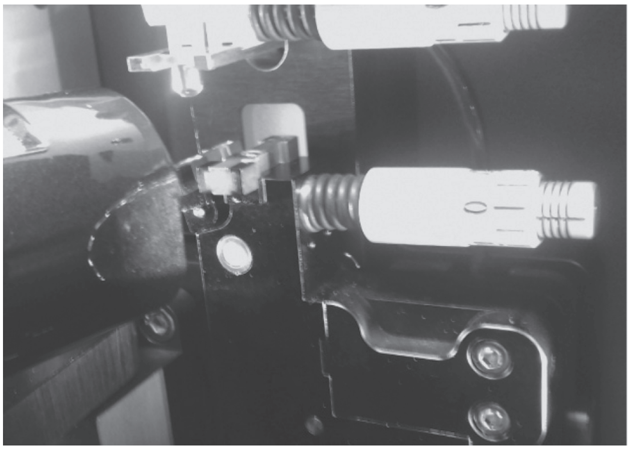

Fig. 3. Photo of parts of a machine with a mounted sample.

pared to the initial LUP carbon tape is a task to be solved.

Another binder was PA-6 highly molecular polyamide (the melting point is 245°C), which is a thermally resistant and thermoplastic material with high adhesion to different, even metal, substrates [5].

First, samples to be pull-out tested were fabricated on a Fimabond installation (produced by Textechno Corp.) (Fig. 1) using suitable crucibles (Fig. 2), the external diameters of which were 4 mm. Prior to measurements, elementary filaments were manually pulled out of LUP carbon tape, each of them was inserted in a special device with an internal diameter of 0.5 mm. Each filament was captured and oriented automatically in this device, because the latter operates as a vacuum cleaner, allowing samples to be made by operators even if they are unfamiliar with the machine. After that, an empty crucible filled manually with a binder was installed onto a special substrate. The accuracy of elementary-filament immersion into a binder was controlled by three cameras installed on a Fimabond system. After immersing the elementary filament into a binder to a depth of 2 mm, the curing process was conducted on the same setup. The average time of sample preparation was 30 min. The use of thermoplastic adhesive enabled one to slightly reduce the time for preparing a sample (to 20 min), because melting was a substitute for curing and no time was thus spent on producing a binder.

The installation procedure of an elementary filament in the molten binder was completely identical: a filament was fixed in a melt, and thermoplastic binder was then cooled to room temperature on a Fimabond setup.

A ready-made sample (see Fig. 2) was positioned in a Textechno Favimat+ breaking machine, as is illustrated in Fig. 3. The setup exhibits high precision and allows one to affix elementary filaments with diameters from 1 to 10 μm in special precision devices. Sam-

Table 1. Properties of an elementary-filament—binder system before and after applying metal coatings onto LUP carbon tape

Parameter	LUP tape			
	initial (coating-free)	coated		
		titanium	steel	titanium + steel
Epoxy binder				
Adhesive strength, MPa	87	83	112	79
Polyamide binder				
Adhesive strength, MPa	37	43	56	40

Table 2. Interlayer-shear strength of carbon-fiber-reinforced polymers

Metallization technology	Interlayer-shear strength, MPa		
	titanium	stainless steel	titanium + stainless steel
Uncoated	35		
Without prepurification	57	54	35
With argon purification for 3 min	63	61	41

ples were loaded at a gripper movement rate of 0.1 mm/min.

The interlayer shear was also examined in this work. For this purpose, standard samples were prepared via vacuum infusion, giving a unidirectional composite material with 25 layers laid out in accordance with a 0 reinforcing scheme. Coatings were applied onto a raw carbon tape and onto a tape subjected to 3 min of the argon exposure. Both tapes served afterward as precursors of samples for interlayer-shear tests. The binder in the interlayer-shear samples was a composite of ED-20 epoxy resin, DETA hardener, and DEG-2 (diethylene glycol) active solvent. Similarly to in the case of pull-out samples, the relative changes in strength compared to the initial LUP carbon tape were preferable to its absolute values. All interlayer-shear and pull-out tests were carried out at room temperature.

RESULTS AND DISCUSSION

Table 1 shows the mean values of adhesive strength of elementary filaments used as precursors for LUP carbon tape with epoxy and polyamide binders. For each sample, the diameter of the elementary filament was measured in the automatic mode at its point of contact with the polymer matrix at an accuracy to 0.01 μm, which enabled the adhesion strength to be evaluated with minimum errors.

It is evident that the adhesive strength of the elementary filament of the LUP carbon tape atop an epoxy binder (with any type of coating) is almost twice that on a thermoplastic adhesive.

The adhesive strength of epoxy binder coated with titanium becomes only 0.3% lower, but declines by 5% when a bilayer coating is used. On the other hand, elementary filaments covered with stainless steel exhibit a 34% gain in adhesive strength.

For a polyamide binder, the above types of coatings led to an increase in adhesive strength by 14.7, 50, and 6.2%, respectively, for titanium, stainless-steel, and bilayer coatings compared to the raw material.

Table 2 includes the interlayer-shear strength values for all inspected samples.

Applying a bilayer coating causes a very slight decrease in strength that coincides with data on strength evaluated through the pull-out tests.

Single-layer coatings favor an increase in the strength of carbon-fiber-reinforced plastics within the interlayer shear. For a titanium coating, the interlayer-shear strength is 61% higher, while it increase by 52% for a stainless-steel coating.

Preliminary argon exposure of the carbon tape before it is coated by the metal layer favors a gain in interlayer shear-strength for all the studied coatings.

CONCLUSIONS

It was established that applying stainless-steel coatings onto a carbon tape led to a gain in adhesive strength at using both epoxy and polyamide binders. On the other hand, titanium and titanium + stainless-steel bilayer coatings in combination with epoxy binder caused a loss in adhesive strength, showing them to be inappropriate materials for the production of carbon-fiber-reinforced polymers.

The strength evaluated via the pull-out tests for a carbon tape coated with a stainless steel was found to be 34% higher than for a raw sample. At the same time, substituting an epoxy binder with a less durable polyamide was shown to improve adhesive strength for all used coatings.

The interlayer-shear tests, similarly to the a pull-out method, revealed an increase in strength by 61% for a tape covered with titanium and by 52% for that coated with stainless steel in comparison with uncoated carbon tape. Since the epoxy binders involved in the fabrication of samples for pull-out and interlayer-shear tests were different, it was inexpedient to make any comparison between them. In any case, applying metal coatings onto a carbon tape resulted in a gain in adhesive strength when using both epoxy and polyamide binders.

ACKNOWLEDGMENTS

This work was supported in the framework of the project "Research on the Development of Composites with a Controlled-Chaos Structure and Their Application in Advanced Technologies" within assignment no. 11.7291.2017/BCh.

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Translated by O. Maslova

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