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Sergei EGOROV, Tatiana TARASOVA, Svetlana TEREKHINA - Production technology for polymeric composite materials by additive manufacturing methods - In: International Conference on Modern Trends in Manufacturing Technologies and Equipment, ICMTMTE 2020, Russie, 2020-09-07 - IOP Conference Series: Materials Science and Engineering - 2020

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Production technology for polymeric composite materials by additive manufacturing methods

S A Egorov^{1,*}, T V Tarasova¹ and S M Terekhina²

¹Moscow State University of Technology “STANKIN”, Vadkovsky lane., 1, Moscow, 127055, Russia

²Ecole Nationale Supérieure d’Art et Métiers ,2 Boulevard du Ronceray, 49100 Angers Cedex 01, France

*sergey_951@mail.ru

Abstract. Additive manufacturing methods make it possible to create complex geometry parts, that cannot be produced by conventional methods. For manufacture of the products, composite polymeric materials can be used, both disperse-filled and filled with short or continuous fibers. Recently, more and more attention has been paid to reinforcing plastics with natural fibers. The study of the manufacturing process of a polymeric composite material filled with continuous natural fibers by the fused filament fabrication is presented

1. Introduction

1.1. Overview

Additive manufacturing (AM) or layer-by-layer technologies of production – one of the most rapidly developed direction of digital manufacturing. AM includes various technologies such as 3D printing, rapid prototyping, direct digital production, layered production [1].

Methods of additive technologies make it possible to create products of complex shapes that cannot be manufactured by conventional methods. These methods help to reduce the time between project design and product manufacturing compared to conventional methods [2].

A wide range of materials available for additive production (from metallic powders to polymer filaments) makes it possible to produce analogues of products obtained by conventional methods, often not inferior to them by mechanical characteristics [3, 4].

At present, all technologies of additive production are fully reflected in the literature [5, 6]. The main attention is paid to the manufacture of products from metal powders [7] using selective laser melting technology, in view of the increased interest in obtaining functional parts of various machines and mechanisms [8].

However, polymeric composite materials (PCM), filled with fibers of various types and composition [9, 10], already constitute a serious competition to metals. In this connection, on the wave of general interest in additive technologies, attempts are made to directly produce composite materials using 3D printing.



1.2. *Materials review*

For manufacture of the products, PCM can be used, both disperse-filled [11] and filled with short or continuous fibers [12, 13].

PCM not only reflect the properties of the matrix and filler, but also have completely new characteristics, which are often not inherent in individual components.

Matrix of PCM, as the name implies, are polymers. The invention of polymers with new sets of properties led to the expansion of the possibilities for their use, which led to a rapid increase in production volumes from polymeric materials. However, the main problem associated with the use of polymeric materials is still their disposal, which involves a sets of complex and expensive processes. The wider use of biodegradable plastics can solve this problem.

According to the American Society for Testing and Materials (ASTM) standard, biodegradable plastics are plastics that decompose as a result of the action of natural microorganisms such as bacteria, fungi. The main advantage of biodegradable polymers is their ability to decompose quickly compared to polymers derived from petrochemical raw.

Despite the wide variety of biopolymers produced, a special place in this sector belongs to polylactide (PLA), a material based on lactic acid. Polycaprolactone (PCL) is also quite widespread. Due to its low melting point (about 60 °C), PCL is used only in medicine (the creation of self-dissolving implants) and in prototyping. Polylactide is widely used in industry.

Products made from polylactide compete in physical and mechanical properties with similar products from ABS plastic, one of the most popular materials both in the field of 3D-printing and in the polymer industry as a whole.

Currently, PCM have found application in almost all industries. PCM are widely used in the manufacture of machine parts and mechanisms, elements of various designs, electrical insulation [14, 15].

In terms of mechanical characteristics (per unit mass), fiber-reinforced polymer composites (FRPC) can significantly exceed traditional structural materials, which can reduce the volume and mass parameters of a product many times over.

The shapes and types of reinforcing fibers are also diverse. At present, fillers made of organic, aramid, basalt fibers, as well as carbon fibers, glass fibers, etc. are widely used for reinforcing composite materials. They are part of the composite in the form of short fibers, ribbons, threads, bundles, fabrics, nonwoven materials and other fibrous structures

The properties of FRPC significantly depend on the reinforcement scheme, as well as on the characteristics of the interaction of components at the interface. To achieve the required characteristics between the fibers and the matrix, a certain ratio of properties must be maintained, high adhesion between them and the wettability of the filler by matrix [16, 17].

Recently, more and more attention has been paid to reinforcing plastics with natural fibers. [18]. Interest in natural fibers is associated with economic factors, since they are a renewable resource, and their cost is low compared to all other fibers. For this reason, natural fibers are promising for use in the manufacture of PCM as reinforcing fillers intended for the manufacture of a wide variety of products not exposed to a humid environment. PCM with this type of fibers has another important advantage - the possibility of processing and reuse. Due to its porous structure, plant fibers are well impregnated with a polymer matrix, which provides high strength at the polymer-fiber interface.

The properties of the most common fibers in the industry are presented in table 1 [19]. There is a superiority of the properties of glass fiber, and especially carbon fiber over natural fibers, however, if we compare the density of materials, and, accordingly, the mass of the product or specific strength, the difference does not look so significant.

Table 1. Properties of different fibers

Fiber	Density, g/cm ³	Fiber diameter, μ m	Break elongation, %	Specific strength, g/tex
Cotton	1,2	11-12	7	0,8
Flax	1,3	5-40	3	1,3
Jute	1,5	8-30	2	0,5
Glass fiber	2,55	5-24	2-5	1
Carbon fiber	1,9	5-7	2	10

1.3. Production methods review

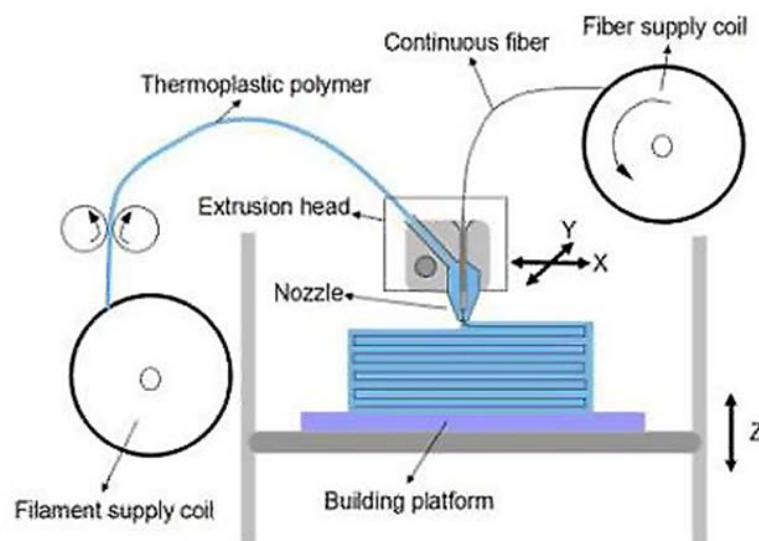
FFF (Fused Filament Fabrication) technology is one of the most widely used 3D printing technologies. Currently, consumer versions of FFF printers with open-source manufacturing modes occupy the widest part of the 3D printer market due to the simplicity of the manufacturing process, flexibility, and the low price of the installations and materials [20]. The most commonly used materials are thermoplastics such as ABS, PA, PLA [21].

The basis of fibrous fillers are elementary fibers (continuous or short). Elementary fibers can be used independently or for the production of other forms of fibrous materials - threads, ribbons, fabrics, canvases, etc.

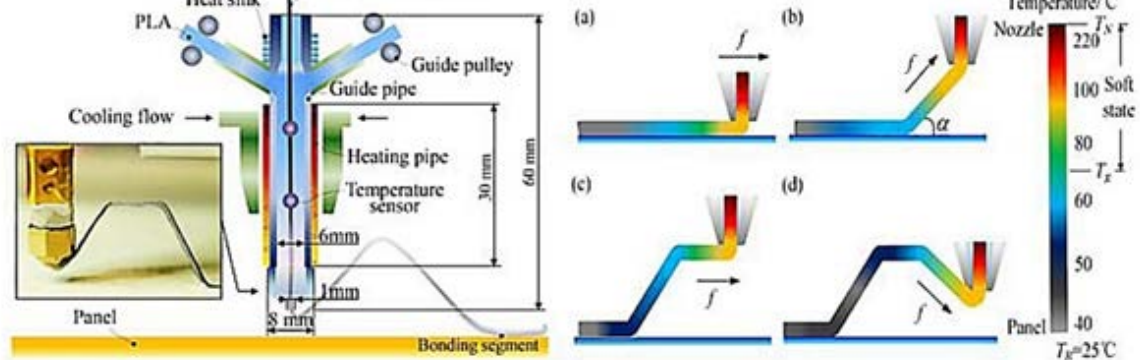
When using additive technologies for the production of FRPC, equipment needs to ensure the integration of the reinforcing component into the matrix during printing

By type, the reinforcing components are divided into continuous and short fibers. With regard to the production of composite material on a 3D printer, it should be borne in mind that continuous fibers have a strong reinforcing effect, however, require laying. Short fibers can be pre-mixed with polymer powder or filament, but after printing they will be oriented randomly (unless orientation operations are provided).

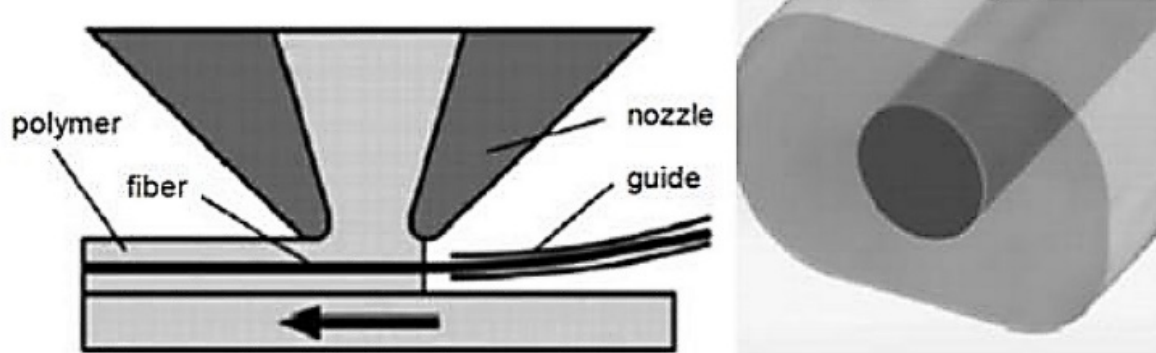
The method of fiber integration is determined by the selected additive technology (FFF). One of the ideas for reinforcing in this process is as follows: in addition to the polymer, fiber is continuously fed through the extruder, which, under the influence of high temperature and pressure in the chamber, is impregnated with the base material and leave the nozzle in a single stream (Figure.1).

**Figure 1.** Manufacturing scheme of FRPC by FFF [22]

Heat sink | Carbon fiber bundle | Temperature (°C)



Authors [24] for the manufacture of FRPC proposed the addition of a fiber feed tube synchronized



It should be noted that attempts to combine additive methods and principles for producing composite

2. Materials and methods

As the basis for the setup, it was decided to use the TEVO Black Widow 3D printer (Figure. 4), and modify it to be able to print with composite material reinforced with continuous natural fibers.

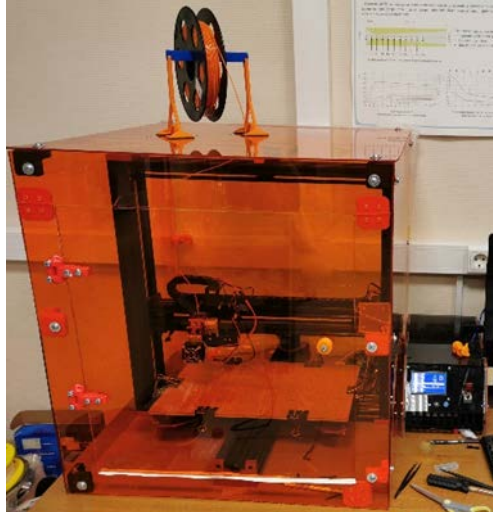


Figure 4. Basic 3D printer TEVO Black Widow

A commercially available thermoplastic was chosen as the matrix material, and a continuous natural fiber with a diameter of 0.2 mm was the reinforcing component.

The mechanical properties of the polymer were tested in the Zwick machine with a 500N force sensor in accordance with the GOST 4648-2014 (ISO 178: 2010).

Tests were conducted for samples made in two directions of printing (XZ and XY), as shown in Figure. 5

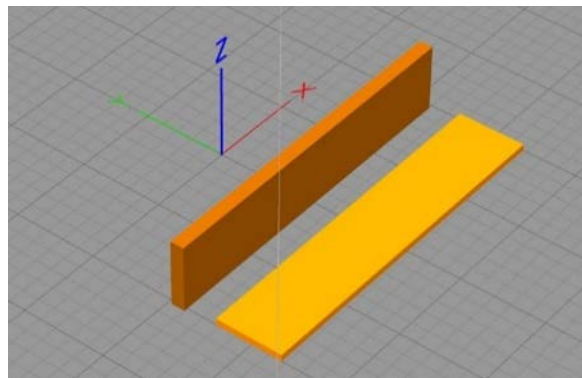


Figure 5. Printed samples in XZ and XY direction

3. Results and discussions

Initially, the mechanical properties of the polymer matrix were investigated. The results are presented in table 2.

Table 2. Mechanical properties of FRPC matrix

	E_{flex} , MPa	σ_y , MPa	σ_{ult} , MPa	ε_f , %
XY orientation	393.91±20.74	10.5±0.54	14.5±0.75	6.2±0.11
XZ orientation	499.83±18.4	13.2±0.45	17.9±0.62	6.24±0.13

Samples of composite material for testing in accordance with ISO 527-4 were manufactured on the above equipment. The image of the sample and the cross section of the monolayer are shown in Figure. 6 a, b

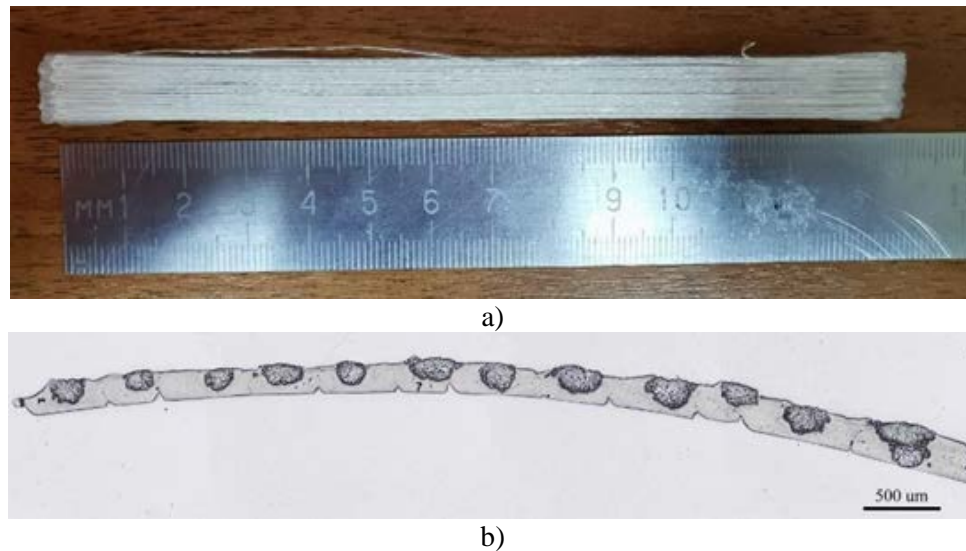


Figure 6. Specimen (a) and the cross section of the monolayer (b)

The porosity of the samples is quite low ($\sim 4\%$), compared with printed samples filled with synthetic (glass and carbon) fibers, the porosity of which reaches $\sim 15\%$ [25, 26].

Rational regimes of material fabrication were found and showed in Table 3.

Table 3. Rational regimes of material fabrication

Parameter	Value
Chamber type	closed
Extruder temperature, $^{\circ}\text{C}$	220
Bed temperature, $^{\circ}\text{C}$	100
Layer height, mm	0,3

Filling degree of obtained composite is 30%. It is close enough to commercially available high-strength carbon fiber filaments with a polymeric matrix [27]. The distance between the filaments within one layer was $\sim 480\ \mu\text{m}$, the layer height $\sim 290\ \mu\text{m}$.

4. Conclusion

The possibilities and prospects of manufacturing polymeric composite materials by the methods of additive technologies are considered.

A study of the manufacturing process of the polymeric composite material filled with continuous natural fibers by the FFF is presented.

A technological process developed for producing a new easily processed polymeric composite material by the FFF with a ratio of components: 70% (by volume) of the polymer and 30% (by volume) of continuous natural fiber.

Future work will be aimed at studying the physic-mechanical properties of the obtained composite material. As well as various options for placing fibers and the development of technology for controlling the degree of filling in various sections of the specimen.

Acknowledgements

The work was carried out with the financial support of the Ministry of Education and Science of the Russian Federation in the framework of state task (project No. 0707-2020-0034).

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