Research of biodegradable wood completed composite materials based on polylactide

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Abstract. Experimental samples of biodegradable composite material based on polylactide and wood filler, subjected to thermal modification, were obtained. When studying the samples a change in such properties as the rate of biodegradation and tensile strength was observed. It has been established that the degree of preliminary thermal treatment of wood filler can control the rate of biodegradation in the material, which can be used in the production of biodegradable packaging. Depending on the desired product life cycle, packages with an adjustable decomposition time can be created. It is found that with an increase in the degree of heat treatment of the filler in a composite material with more than 40 % of polylactide the tensile strength decreases, while with a lower binder content thermal modification has a positive effect on the strength. Also, adding the fillers can significantly reduce the cost of composite material.

1. Introduction

Currently, the environmental situation in the world is rather unfavorable. A constantly growing population requires more and more food, consumer goods and housing. All this leads to the growth of cities, industry and an increase in the volume of municipal solid waste, among which polymers have the greatest environmental damage. So, decomposition of office paper in the natural environment requires up to 2 years, a tin can up to 10 years, a rubber cover up to 120-140 years and a plastic bottle about 180-200 years. Today, the share of plastic waste worldwide is growing at an alarming rate. At the moment, they occupy third place in the ranking of the main factors polluting the Earth. Synthetic polymers, such as polypropylene, polyethylene, polyvinyl chloride and polyethylene terephthalate are highly resistant to solar radiation, temperature, water and microorganisms [1]. Disposal by pyrolysis or incineration also does not significantly improve the environmental situation. In certain degree this issue is solved by recycling, but it requires considerable energy and labor costs.

So today, the prevention of environmental pollution by synthetic polymers, the decomposition of which in natural conditions is much higher than the human lifespan, is the urgent issue. A solution to this problem is the development, manufacture and application of polymers, capable of biodegrading relatively quickly (from several months to several years) under appropriate conditions.

One of the most promising polymers in this direction is polylactide – aliphatic polyester-based hydroxycarbonate acids. The raw material bases for its production are renewable sources of plant origin. PLA is characterized by thermoplasticity, allowing it to be easily processed, as well as the rate of decomposition in the biological environment, while not having harmful effects on it [2]. Due to these properties, polylactide can be used to create new materials of interest not only from the...
standpoint of environmental protection, but also possible applications in medicine, packaging and food industries, as well as in agriculture and in the home.

However, the cost of production of polylactide today remains quite high, which is an inhibiting factor for the development of bioproduction. In this regard, to reduce the cost of production of composite materials and end products based on PLA, efforts have been made by mixing polylactide with vegetable filler in the form of wood flour subjected to thermal modification [3-4]. The addition of thermomodified wood flour can be justified by the following criteria. Firstly, thermal modification allows removing hemicellulose from wood, which adversely affects the contents of packages, reducing the shelf life of products. Secondly, thermal modification imparts some properties to wood, such as high water resistance, hardness, biological and chemical resistance. Thirdly, thermally modified wood is environmentally friendly, which is important for food packaging materials. Also the use of such filler will allow efficient utilization of wastes of woodworking industry [5-6].

2. Materials and methods

The samples of polylactide composites with the contents of both heat-treated at $t = 180-200^\circ C$ and untreated wood filler of brand 140 were prepared for the study of physical and mechanical characteristics. Polymer polylactide granules 4043D were used as a binder. The quantitative composition of the samples and the processing temperature of the filler are given in table 1.

| Sample, No. | PLA, % | Untreated wood filler, % | Wood filler treated at $t=180^\circ C$, % | Wood filler treated at $t=200^\circ C$, % |
|-------------|--------|--------------------------|------------------------------------------|------------------------------------------|
| 1           | 100    | 0                        | 0                                        | 0                                        |
| 2,3,4       | 70     | 30                       | 30                                       | 30                                       |
| 5,6,7       | 50     | 50                       | 50                                       | 50                                       |
| 8,9,10      | 30     | 70                       | 70                                       | 70                                       |

Initially, blanks for samples were obtained by mixing the components at a temperature of $180^\circ C$ for 5 min using the equipment of Brabender GmbH & Co KG. Further, these blanks were placed in a Gotech GT-7014-H pressing machine at a temperature of $185^\circ C$ for 5 minutes, and then they were cooled for 2 minutes. The photos of the obtained samples are presented in figure 1. Next, samples of the same size were cut from the blanks for further testing.

**Figure 1.** Samples of the composite material: 1 – pure PLA; 2 – thermally untreated filler; 3 – filler is heat treated at $180^\circ C$; 4 – filler is heat treated at $200^\circ C$.

To determine the ultimate tensile strength of composites, the tensile testing machine JLTTC LDS-5L was used (figure 2).
3. Results and discussion
In the course of studies it is revealed that the destruction of composite samples in compost occurred in a two-step stage. In the initial phases of degradation the chains of high molecular weight polylactic acid were hydrolyzed to oligomers with a lower molecular weight. That reaction depended on temperature and humidity. Moreover, it could be accelerated by hexone bases or acids [7–8].

Fragmentation of the composite earlier occurs on the samples filled with wood filler. In this case, heat treatment of wood filler reduces the rate of biodegradation of the composite. Figure 3 presents experimental curves showing the destruction time of polylactide and composites based on it.

![Figure 3](image)

Figure 3. Destruction of composites depending on the type of wood filler.

The experimental results clearly show that the biodegradability of the PMC component in a composite of pure polylactide without the addition of wood filler is significantly lower than that of composites filled with wood. In this case, the destruction of the composite with thermally treated filler (200 °C) was lower than that of a composite with thermally untreated filler [9–10].

Utilization of biodegradable polymer can be cheaper, technologically easier and be done by degrading it in alkaline aqueous solutions of NaOH and KOH. Such a disposal method can simulate an accelerated process of PLA degradation in real conditions. The composites in the form of thin plates subjected to destruction in aqueous solutions of NaOH and KOH were studied in this article. The temperature of the solutions was 40 °C and 60 °C. Every 10 minutes the samples were removed from aqueous solutions of alkalis and weighed to detect weight loss. This procedure was repeated until the samples were completely destroyed. The degradation time of the polymer at 40 °C and 60 °C in these solutions is presented in figures 4 and 5. Studies have shown that the time of degradation of polylactide was independent of concentration and alkaline solution. The difference in PLA degradation time at 60 °C is greater than at 40 °C. In this case, with an increase in temperature by 20 °C, an increase in the rate of degradation of polylactide by several times is observed. At a temperature of 60 °C, a solution of potassium hydroxide accelerates the degradation of the polymer by about 1.5 times compared with sodium hydroxide. It is important to note that the concentration of
solutions does not affect the total destruction time of the samples. After analyzing the data obtained, we can conclude that the rate of degradation of polylactide in alkali solutions is more dependent on the temperature of the alkali solution. Therefore, when creating a technology for the disposal of composites and materials from PLA by degradation in alkaline solutions, the process temperature should be considered first [11].

![Figure 4. Degradation time of polylactide at 40 ºC.](image1)

![Figure 5. Degradation time of polylactide at 60 ºC.](image2)

In addition to biodegradation, the samples were investigated in tension. According to figure 6, an increase in tensile strength was observed in samples with filler content within 30 %. At the same time, samples with a high content of wood filler (50 % and 70 %) tend to decrease the tensile strength. This is explained by the fact that with an increase in the number of wood particles in the sample, the adhesive component between the polymer and the wood filler decreases (due to the low orientation of the filler particles) [12]. Moreover, it should be noted that for samples from pure PLA the tensile strength is in the range of 50.04 N / mm².

![Figure 6. The dependence of the tensile strength on the temperature of the processing of wood filler.](image3)

According to the histogram (figure 7), the samples with the lowest content of filler have the maximum strength values in terms of the amount of wood filler content.
Figure 7. The histogram of the dependence of the tensile strength on the content of wood filler.

4. Conclusion
In the course of investigations it was revealed that the use of plant materials as filler increases the biodegradable properties of the composite material. Preliminary heat treatment of the filler slows down the process of decomposition of the composite compared to composites containing thermally untreated filler. The studies of the strength of composites in the gap have also been conducted. It is found that there is an increase in tensile strength of the samples with filler content of 70 %. At the same time, there is a tendency to decrease in strength in samples with lower content of wood filler (50 % and 30 %). It is assumed that depending on the further purposes of the application of the composite, focusing on the results obtained, it will be possible to set qualitative and quantitative composition.

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