Technology development for the production of thermoplastic composites with agricultural fillers by compounding method on co-directional twin screw extruder

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Abstract. The paper is devoted to the development of a continuous process for the production of thermoplastic composite materials based on low-pressure polyethylene and agricultural plant waste products by the components compounding on a Rheomex PTW 16 PolyLab laboratory twin-screw extruder. The processing modes of the developed compositions are given. The effect of melt degassing on the quality of the obtained granulate is noted.

Keywords: continuous process technology, thermoplastic composite, polyethylene, agricultural origin fillers, dispenser, compounding, twin-screw extruder, degassing, vacuum treatment, granulate.

1. Introduction
The main problem for thermoplastic composite materials with agricultural origin filler creating is the problem of uniform distribution of components by mass when they are mixed and compounded. This process in practice is carried out firstly by periodical method in rotary and roll mixers and secondly by continuous one in single-screw pin extruders and co-directional twin-screw extruders. Recently, to obtain the specified class of composite materials, twin-screw extruders of co-directional rotation are more often used [1, 2]. They provide high-quality high-speed compounding of components and obtain granulate composites with high uniformity of components distribution.

The mixing effect in twin screw extruders is provided by screws located parallel to the cylinder and rotating towards each other or in the same direction. In the opposite rotation of the screws, they work like rollers, while the material is fed along the screws. The shift and grinding of the material occur both between the turns of the screws and in the gap between the screws and the cylinder body. The value of material reverse flow increases with increasing distance between the screws and enhances the mixing effect. When the screws rotate in one direction, if they fully engaged, large shear deformations are achieved. It allows to mix highly viscous materials with high content of fillers [3].

A number of researchers [4–7] speak of advantages of twin-screw extruders in the process of compounding thermoplastic polymers with various modifiers. Due to their high productivity and good mixing properties, twin-screw machines can be used to implement a direct (single-stage) extrusion scheme of composites, in which the mixing of the components and the extruding of the resulting compound through the spinneret is carried out as a single stage. In this case, the extruders should be equipped with an effective degassing device (vapors and gases removal) and have wear-resistant surfaces.

The two-stage process for the production of wood-polymer composites consists of the pre-production of compound granulate with its subsequent processing by heat pressing, and injection molding or extrusion.
The possibility of processing composite materials with a thermoplastic polymer matrix and lignocellulose fibers on a twin-screw extruder was shown in this study [8]. It was noted that the particle size and fiber length significantly affect the compounding process.

Compositions of polypropylene and sunflower husk with organoclay modification were studied in this study [9]. Compounding of the components was carried out on a Leistritz Micro-18/GL-40D twin-screw extruder. It has been shown that the introduction of a small amount of modifier such as organoclay (5%) contributes to the improvement of the mechanical characteristics of composites. This improvement is due to the uniform distribution of particles in the polymer matrix, which has been proved by a scanning electron microscope using.

The purpose of this experimental study was the laboratory testing of a continuous process for producing thermoplastic composites filled with barley straw and wood flour on a twin-screw extruder with co-directional rotation, as well as studying the physical and mechanical characteristics of the obtained composites.

2. Materials and Methods
To obtain the composite material, such filler was used as:
- barley straw with a dispersion of 200 ... 350 microns, a moisture content of 8.7% and a bulk density of 0.14 g/cm³;
- wood flour with a dispersion of 180 ... 250 microns, humidity of 7.9% and bulk density of 0.16 g/cm³.

Grinding of agricultural origin fillers was carried out on a rotary knife mill. Humidity was determined by thermogravimetric method by a moisture analyzer "Evlas-2M".

Low pressure polyethylene (HDPE 273-83) manufactured by Kazanorgsintez according to GOST 16338-85 was used as the polymer matrix.

As a result, samples of compounds of two compositions were obtained:
composition 1: 55% HDPE + 45% barley straw;
composition 2: 55% HDPE + 45% wood flour (powder).

Compounding was carried out on a Rheomex PTW 16 PolyLab twin-screw extruder. The appearance of the extruder with peripheral devices is shown in Figure 1.

The line includes the HAAKE RheoDrive 7 OS, the Rheomex PTW 16 PolyLab measuring extruder-compounder (modular 16 mm twin-screw co-directional extruder with an L/D ratio of 40:1), a cooling bath, a granulator and a metering system including volumetric single-screw DRS 28 for polymer granules, MT 1 volumetric twin-screw metering unit for fibrous fillers, Vertical Stuffer Feeder screw feeder for powder fillers, vacuum pump and Accel 500LC circulation cooler.

The equipment starts as follows.
Figure 1. Appearance of a twin-screw extruder-compounder Rheomex PTW 16 with peripheral devices: 1 - extruder body; 2 - vacuum pump; 3 - agricultural origin fillers dispenser; 4 - boot device; 5 - polymer granulate dispenser; 6 - supply of powder filler; 7 - heated case with screws; 8 - adapter of the degassing system; 9 - a cooling bath with a temperature control system; 10 - thermostat; 11 - extrudate (strand) with air cooling; 12 - device for air drying and strand cooling; 13 - granulator

The cylinder was opened and the screws were cleaned of a protective lubricant with detergent. Then, the extruder heating zones were turned on and the temperature controllers were installed at the required temperatures (PolySoft OS program). The extruder reached operating temperatures within 10 minutes. By turning it on at a low speed (5 rpm), it was found that the screws rotate freely. Next, the PolySoft OS program and the drive unit were turned on, then, with free rotation of the screws at a frequency of 50 rpm, dispensers were switch on, which began to feed components (polyethylene granules and agricultural origin fillers) into the loading device of the extruder. Further, the screw speed was increased to the recommended value at which the torque on the extruder drive does not exceed 60% of the maximum permissible value (130 Nm).

At the first stage, the masterbatch “HDPE 273-83 + agricultural origin fillers” has been obtained. Based on the metering calibration schedules, the relative rotational speeds of the metering screws were determined. For the MT 1 dispenser (due to its low productivity), the maximum (100%) rotation frequency was set, the rotational speed of the DRS 28 dispenser was calculated based on the ratio of filler to polymer.

The processing modes of thermoplastic composites are shown in Figures 2-3.
Figure 2. The processing mode of the composition “55% HDPE 273-83 + 45% wood flour”

Figure 3. The processing mode of the composition “55% HDPE + 45% barley straw”
Figures 4-6 present some features of the granulation process.

Figure 4. The output of the strand (extrudate) from the extrusion head into the cooling bath

Figure 5. The output of the strand (extrudate) from the cooling bath to the drying device

Figure 6. The output of the strand into the granulator (air cooling of the extrudate)

To carry out physical and mechanical tests by hot pressing on a Gibtiré hydraulic press, plates with a size of 200×200×1 mm were formed, from which the plates were cut.

The density of the composites was determined on an H-200L densitometer with an ultrahigh resolution of 0.001 g/cm³ according to GOST 15139 - 69.

The values of water absorption of the samples were determined according to GOST 4650 - 80 (method A, water exposure for 24 hours).

Physical and mechanical tests of the prepared samples were carried out on a UAI-7000 M tensile testing machine at a temperature of 23 ± 2°C and a clamp speed of 1 mm/min. The tensile strength and
tensile modulus were calculated according to GOST 11262 - 80 and GOST 9550 - 81. The measurement error for all tests was not more than 10%.

3. Results and Discussion

Table 1 shows the physical and mechanical characteristics of the obtained thermoplastic composites.

| Composites | Tensile Strength, MPa | Tensile modulus, MPa | Relative extension, % | Moisture absorption, % | Density, g/cm³ |
|------------|-----------------------|----------------------|-----------------------|------------------------|---------------|
| Composite 1 | 13,5                  | 1830                 | 7,4                   | 5,1                    | 1,15          |
| Composite 2 | 12,8                  | 1750                 | 6,9                   | 5,3                    | 1,12          |

Figure 7 shows typical tensile curves for the resulting thermoplastic compositions.

![Figure 7. Typical tensile curves of thermoplastic composites filled with 45% wood flour (a) and 45% barley straw (b)](image)

The values of physical and mechanical characteristics shown in Tab. 1 and Figure 1 are consistent with the data of other researchers, which gives reason to believe that it is possible to use fillers from wood flour and barley straw to obtain composite materials for engineering purposes.

Thermoplastic composites with such fillers should, if possible, be processed without preliminary drying, but the resulting products should meet high operational and technological requirements. To a large extent, this set of requirements depends on the porosity of the initial granulate, the reason for which is air capture, moisture in the feedstock, as well as partial decomposition of thermoplastics and agricultural origin fillers under high temperatures in the extruder with the release of volatile substances. In practice, this means that gas inclusions must be removed from the extruder before the forming head.

To combine extrusion with the operation of removing volatile substances released during processing, the extruders should be equipped with devices for melt degassing. The gaseous components are sucked off in all cases in the areas of expansion of the molten mass through holes in the cylinder of the extruder [1, 3]. An extruder Rheomex PTW 16 is equipped with a similar device such as vacuum pump 2 and adapter 8 (Figure 1).

Figure 8 shows micrographs of a slice of composite granules obtained with melt degassing and without degassing in extruder.
As follows from Figure 8, the granulate obtained without degassing does contain pores and cracks, leading to a decrease in physical and mechanical properties and moisture resistance of the products.

![Figure 8 Photos of the structure of transverse sections (200-fold increase) of the masterbatch compound granulate obtained without degassing (a) and with degassing (b) of the last extruder mixing zone](image)

4. Conclusions

Thus, in the course of the research, a two-stage technology for producing granulate of thermoplastic composite materials filled with wood flour and finely dispersed barley straw by continuous compounding of components on a twin-screw extruder of co-directional rotation was developed.

As a result of the degassing processes study, it was found that degassing of the melt of the compound with a vacuum value of 0.8 ... 0.9 bar can significantly reduce the porosity of the granulate.

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