STUDY OF RHEOLOGY PROPERTIES OF POLYMER COMPOSITE MATERIALS

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ABSTRACT

This article presents the results of the study of the rheological properties of polymer composite materials based on glass microgranules containing high-density polyethylene. The flow curves of the obtained compositions were obtained by the capillary viscometry method. On the basis of the conducted research, simple mathematical models were built, which allow to estimate the viscosity values of the alloys based on the given filler content.

Keywords: polyethylene, glass microgranules, polymer composite materials, rheological properties.

Introduction

Polymer materials containing various dispersed fillers are currently widely used for the production of products required for technical or other purposes. One of the most important technological indicators of melts of processed compositions is the rheological properties, especially the effective viscosity coefficient. The need to evaluate the value and determine the patterns of its change in connection with the application of the dispersed filler makes this problem very relevant. For this purpose, glass microgranules were chosen as a dispersed filler. This type of dispersed filler is used to enhance the performance properties of polymer composite materials.

Microgranules have high hardness, smooth surface and are well distributed in polymer matrices. YSP with glass microgranules has high hardness, strength and serviceability. Polymers are characterized by non-Newtonian rheological behavior, but the introduction of a dispersed filler into them causes the flow characteristics of compositions based on them to be complicated. In this case, ideas about the structure of dispersed systems are used to describe the rheological properties [1]. The spherical shape of the filler particles is a simplifying factor that allows determining the effect of the filler composition on the rheological properties of polymer materials filled with particles. Therefore, this type of composite materials is of great interest to researchers [2].

It is known that the rheological properties of high-density polyethylene are related to their structure, which can be considered from the point of view of lattice models and characterized by such parameters as the shape and size of particles, their packing density, and the coordination number of particles.

In addition, there is also a generalized parameter called the fraction of the polymer matrix phase (Θ), which forms a layer between the filler particles [3].

This parameter is calculated using the volume fraction (\Box) and volume of the filler:

According to Θ , high-density polyethylene is classified into five groups:

- 1. diluted systems $(1.0 \ge \Theta \ge 0.9)$;
- 2. undercharged systems $(0.9 \ge \Theta \ge 0.75)$;
- 3. medium filled systems (0.75 $\ge \Theta \ge 0.2$);
- 4. highly charged systems $\geq \Theta \geq 0$);
- 5. overcharged systems ($\Theta < 0$) [3].

In the presented article, the rheological properties of high-density polyethylene corresponding to the first three classification groups were studied.

Research facilities

BorPure MB6561 injection molded type of high density polyethylene (density 0.955 kg/m3) manufactured by Borealis AG of Austria was chosen as the polymer matrix. Measured at 190°C and 2.16 kg load, this brand has a melt flow rate of 1.5 g/10 min.

ShSO-30 brand glass microbeads produced in the Russian Federation were used as fillers. This filler is a spherical solid particle with an average diameter of 30 microns, an actual density of 2400 kg/m3 and a bulk density of 1370 kg/m3.

Preparation of compositions

Compositions made on the basis of high-density polyethylene with glass microgranules were produced in a BRABENDER plastograph with a working chamber of 52 cm3 at a temperature of 210°C. After the polyethylene granules loaded into the heated chamber were completely melted, microgranules were gradually added to them. Rotating shaped rotors mixed the components and formed high density polyethylene with high homogeneity inside the chamber. The mixing time was 10 minutes.

Research methods

Using capillary viscometers, the rheological behavior of BorPure MB6561 polyethylene and its base of high-density polyethylene filled with glass microgranules was studied. The MV-3M device made it possible to determine the speed of the melt flow.

The use of these devices made it possible to obtain more complete information about the rheological properties of the melts of the studied materials at a temperature of 210° C. Capillaries with 1 mm diameter and 2 mm length to hole diameter ratio 1/d=10 and

20 were used for the measurements to account for the input effect.

Results and its discussion

In order to correctly establish experimentally obtained flow curves, it is necessary to determine the inlet pressure losses that occur during the flow of high-density polyethylene melt at the inlet of the capillaries [3]. Graphs for the calculation of input losses represent the dependence of the pressure (Pin) at the input to capillaries of the same diameter on the ratio of the length to the diameter at constant values of the volume flow (Q).

Figure 1 shows the dependences of PBx l/d for the composition with polyethylene and microsphere content of 0.5 rpm obtained in capillaries with a diameter of 1 and 2 mm at Q = $6 \cdot 10$ -10 m3/s. The pressure loss value Δ Ppot is determined by the intersection of the Rvx axis and the plot of Rvx versus l/d as l/d tends to 0.

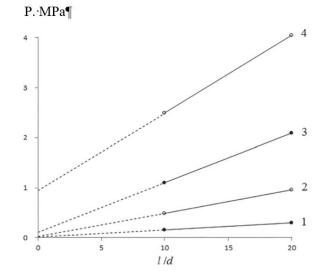


Fig. 1. PBX - 1/d dependence of polyethylene and its composition. 1, $2 - \phi = 0$ rpm; 3, $4 - \phi = 0.5$ rpm; 1, 3 - d = 2 mm; 2, 4 - d = 1 mm

Fig. 2 shows the values of the pressure loss at the inlet of the capillaries at the volume flow rate of the melt $Q = 6 \cdot 10 - 10 \text{ m}3/\text{s}$, depending on the composition of the filler.

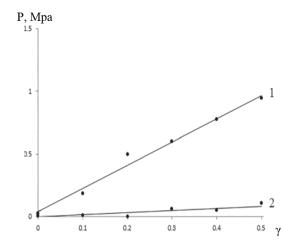


Fig. 2. Dependence of the pressure loss at the capillary entrance on the content of microgranules in the compositions; 1 - d=1 mm; 2 - d = 2 mm.

As can be seen from Figure 2, the pressure loss in capillaries with a diameter of 1 mm was 0.02-0.95 MPa, which corresponds to 2.5-25% of the total pressure drop, while in capillaries with a diameter of 2 mm, the pressure loss was 0. 1 has been. MPa (7% of total pressure).

At higher melt flow rates (Q = 10-9 m3/s and $2.5 \cdot 10-9 \text{ m3/s}$), the entry losses for 2 mm diameter capillaries are also within the specified range. These materials require more energy to melt at the inlet of a smaller diameter capillary than to reorganize the flow rate through the capillary.

Since the materials used in this study are non-Newtonian fluids, the Rabinovich-Mooney correction was applied to construct the flow curves [4].

Fig. 3 shows the flow curves of the materials obtained in capillaries with a diameter of 2 mm at a temperature of 210°C.

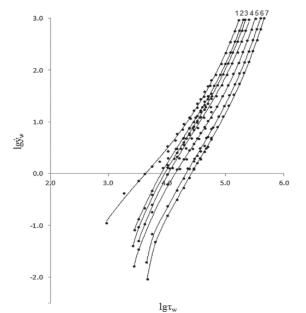
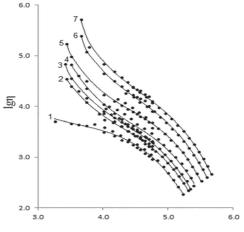


Fig. 3. Flow curves of polyethylene and its base of high-density polyethylene filled with glass granules. 1 - $\varphi = 0$ rpm; 2 - $\varphi = 0.05$ rpm; 3 - $\varphi = 0.1$ rpm; 4 - $\varphi = 0.2$ rpm; 5 - $\varphi = 0.3$ rpm; 6 - $\varphi = 0.4$ rpm; 7 - $\varphi = 0.5$ rpm.

The type of dependencies obtained for filled compositions differs from the corresponding dependency for unfilled polyethylene. At relatively low shear stresses, they are clearly biased toward low shear rates.

Figure 4 shows the dependence of the viscosity coefficient on the shear stress of polyethylene and glass-filled PCM based on it.



 $lg\tau_w$

Fig. 4. Dependence of the viscosity coefficient on the shear stress of polyethylene and glass-filled PCM based on it.

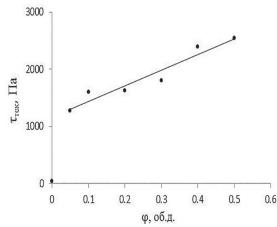
1 - $\phi = 0$ rpm; 2 - $\phi = 0.05$ rpm; 3 - $\phi = 0.1$ rpm; 4 - $\phi = 0.2$ rpm; 5 - $\phi = 0.3$ rpm; 6 - $\phi = 0.4$ rpm; 7 - $\phi = 0.5$ rpm

Such behavior is consistent with the appearance of properties of Bingham (or nonlinear viscoplastic) fluids in compositions, and in this case it is necessary to introduce yield strength as a rheological indicator.

The yield of materials appears at low values of shear stress. The MV-3M viscometer, a device that provides constant pressure in the working cylinder and accordingly creates a constant shear stress, allows you to obtain suitable measurement modes. The value of the output voltage can be estimated using the Casson equation [1]:

$$\tau 0.5 = (\tau \text{tek}) \ 0.5 + (\eta \cdot \gamma) \tag{1}$$

Figure 5 shows the dependence of productivity on the composition of microspheres in polyethylene without filler.





Melting polyethylene without filler does not demonstrate the presence of a yield point. The introduction of a small amount of microspheres into polyethylene (diluted system) leads to the appearance of an increased yield stress with increasing filler content.

Description of rheological properties

To describe the dependence of the shear rate on the shear stress of the compositions, the power law model (2) was used, which allows determining the dependence of the viscosity coefficient on the flow conditions.

$$c = k \cdot \gamma n$$
 (2)

Here, k is the consistency coefficient, n is the flow index or pseudoplasticity index.

The Ostwald-De Villa equation given above is the simplest and most popular model that allows determining the dependence of the viscosity coefficient on flow conditions.

In order to describe the rheological properties of the studied materials, an analysis of the influence of the volume fraction of the filler on the dependence of the effective viscosity

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coefficient on speed and shear stress was carried out. It was found that when the relative viscosity (η rel) is determined at a constant shear stress, the simple Arrhenius model [3] adequately describes the experimental data:

$$\eta rel = eK \cdot \phi \qquad (3)$$

Here, K is the tangent of the slope of the linear approximation of the lnnrel-dependence.

Fig. 6 shows the graph of the natural logarithm.

At constant shear stresses in the range of $6.3 \div 100$ kPa, the relative viscosity of materials from the composition of microspheres is high. As can be seen from the data in Fig. 7, this dependence in the specified range of shear stresses is depicted by a straight line with a slope of K = 5.45. This value is quite close to the values given in [3].

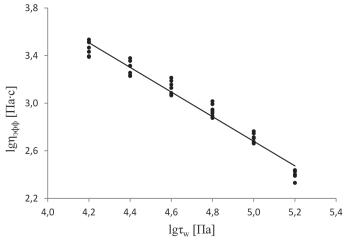


Fig. 6. Dependence of effective viscosity of compositions on shear stress.

Thus, the function (4) can be applied to describe the dependence of the effective viscosity and to fit the compositions:

 $lg\eta comp = log\eta PE + K \cdot \phi$ (4)

Here, ηPE is the coefficient of the effective viscosity of polyethylene at a given shear stress, the coefficient of the Arrhenius model given the logarithmic scale K (K/ln10).

The calculation result gives one generalized curve for all compositions.

Results

Using the capillary viscometry method, the rheological properties of melts of composite materials based on high-density polyethylene filled with glass microgranules were studied. The obtained results made it possible to establish the generalized dependence of the effective viscosity of the compositions on the shear stress and the content of the filler.

In studies, glass microgranules were diluted on the basis of high-density polyethylene, the dependence of the relative viscosity of low and medium filled polymers on the filler content was well described by the Arrhenius model.

The dependence of the flow resistance of the investigated compositions on the content of the dispersed filler was determined.

It was found that the pressure loss at the inlet of the capillary during the flow of the investigated compositions depends on the diameter of the capillary and increases with its decrease. An increase in the content of the dispersed filler in the composition leads to an increase in the pressure loss at the inlet of the capillary.

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POLİMER KOMPOZİT MATERİALLARININ REOLOGİYA XÜSUSİYYƏTLƏRİNİN ÖYRƏNİLMƏSİ

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XÜLASƏ. Bu məqalədə tərkibində yüksək sıxlıqlı polietilen olan şüşə mikroqranullar əsasında polimer kompozit materialların reoloji xassələrinin öyrənilməsinin nəticələri təqdim olunur. Alınan kompozisiyaların axın əyriləri kapilyar viskozimetriya üsulu ilə alınmışdır. Aparılan tədqiqatlar əsasında verilmiş doldurucu tərkibinə əsasən ərintilərin özlülük dəyərlərini qiymətləndirməyə imkan verən sadə riyazi modellər qurulmuşdur.

Açar sözlər: polietilen, şüşə mikroqranullar, polimer kompozit materiallar, reoloji xassələr

ИЗУЧЕНИЕ РЕОЛОГИЧЕСКИХ СВОЙСТВ ПОЛИМЕРНЫХ КОМПО-ЗИЦИОННЫХ МАТЕРИАЛОВ

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АННОТАЦИЯ.

В статье представлены результаты исследования реологических свойств полимерных композиционных материалов на основе стеклянных микрогранул, содержащих

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полиэтилен высокой плотности. Кривые течения полученных композиций были получены методом капиллярной вискозиметрии. На основе проведенных исследований построены простые математические модели, позволяющие оценить значения вязкости сплавов исходя из заданного содержания наполнителя.

Ключевые слова: полиэтилен, стеклянные микрогранулы, полимерные композиционные материалы, реологические свойства.