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Injection Molding of Products Made of Particulate-Filled Polymer Composite Materials with Various Structures

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Abstract—It is shown that the injection molding of thin-walled products with a thickness of less than ~1 mm can be difficult in the case of particulate-filled polymer composite materials (PFPCMs) with MFS-1, MFS-2, and HFS types of structure. Products with a wall thickness of more than ~3 mm and $L/h \leq 100$ can be obtained by injection molding using almost all PFPCM structure types (DS, LFS, MFS-1, MFS-2, and HFS) at the processing temperatures. However, when the L/h ratio increases, the products may be difficult to process using injection molding. For the first time, dependences are established for pressure losses at injection molding on generalized parameters and PFPCM dispersion structure type and product size. Such an approach makes it possible to predict and choose the required process parameters and injection molding machines for processing the majority of PFPCMs with different types of dispersed structures based on polymer matrixes and solid dispersion fillers.

Keywords: polymer composite materials, injection molding, dispersion structure parameters, particulate filler, polyethylene

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Molded particulate-filled polymer composite materials (PFPCMs) lead the technology of processing and obtaining products with a controlled complex of technological and operational properties [1–7].

Injection molding is a common method of PFPCM processing. During injection molding, several processes occur simultaneously connected with deformation in wide temperature and velocity ranges at high pressures with the development of plastic and highly elastic deformations, as well as destruction and cooling with the formation of a dispersed structure of PFPCM.

Control of injection molding technological parameters and use of fillers with various natures, shapes, particle sizes, and concentrations results in changing viscosity of melts and thermophysical properties, which could complicate processing and its mathematical description.

When solid fillers are embedded in polymer matrixes, the viscosity of PFPCM grows, and its dependence can be described by the known Mooney equation up to a certain filler content [7–9]. However, the correlation between viscous properties and dispersed structure of PFPCM, as well as its description in generalized and reduced parameters, cannot be found using the Mooney equation.

For the first time, the correlation of rheological properties of PFPCMs with generalized and reduced parameters and different types of disperse structures was established in [10]; the following types of disperse structures were considered according to the classifications based on structural principles: diluted (DS), low-filled (LFS), medium-filled (MFS), and highly filled (HFS).

This study presents novel data on the filling of injection molds for products with different thicknesses and length-to-thickness ratios with PFPCMs of various dispersed structures. In addition, the optimization of the choice of brand of injection molding machine for the technological process is considered.

PFPCM composites based on low-density polyethylene (LDPE) of 10803-020 grade and glass microspheres of ShSO-30 grade were studied. The prepared composites had different structures and filler contents (φ_f, vol pts) of 0.05 (LFS), 0.12 (MFS-1), 0.2 (MFS-1), 0.27 (MFS-2), 0.32 (MFS-2), 0.34 (MFS-2), 0.39 (HFS), and 0.44 (HFS).

The content of glass microspheres for each structure type was calculated by the following formula:

$$\varphi_f = (1 - \Theta)\varphi_m; \qquad (1)$$

where φ_m is the maximum filler fraction in PFPCM, vol pts.

The maximum content of the particulate filler (φ_m, vol pts) was determined using standard techniques [11]. It was considered as 0.50 vol pts for ShSO-30.

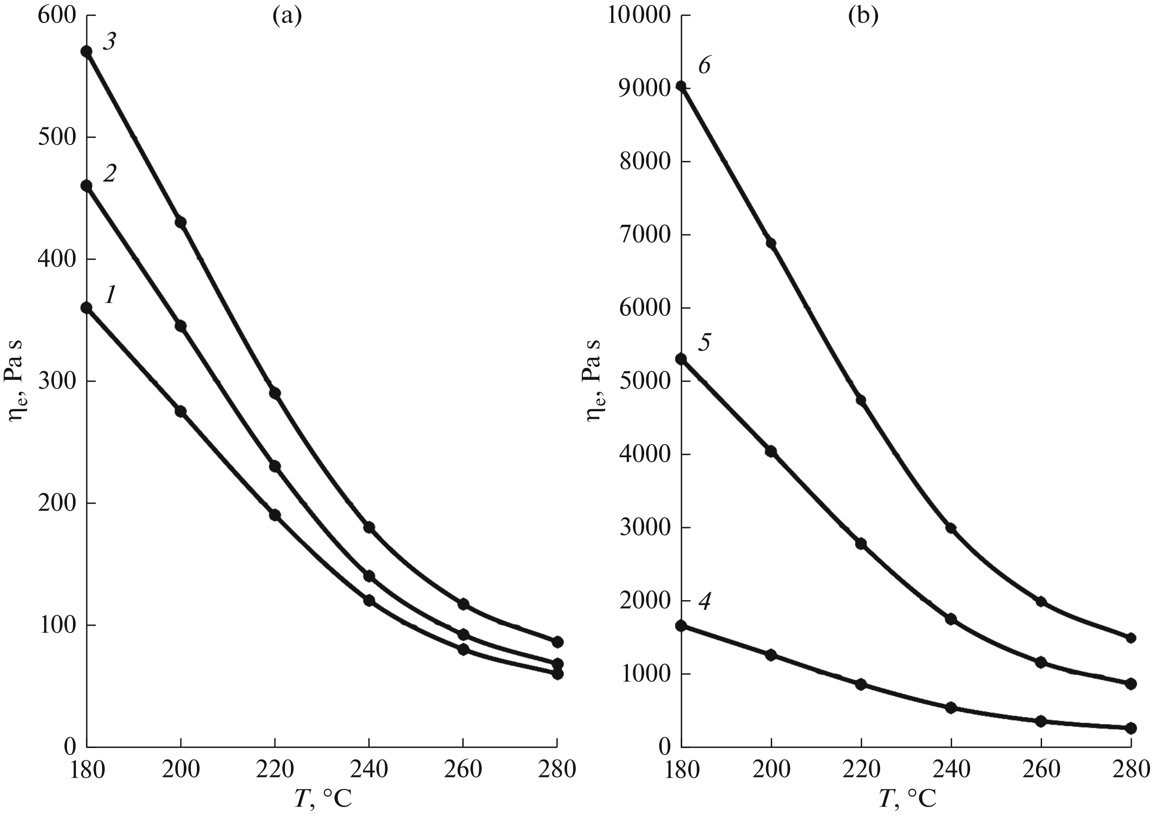

Fig. 1. Dependences of effective viscosity η_e of PFPCM based on LDPE + ShSO-30 on the processing temperature at various values of generalized parameters Θ (vol pts) and structure type: (DS, *1*) 0.98, (LFS, *2*) 0.9, (MFS-1, *3*) 0.75, (MFS-2, *4*) 0.45, (HSF, *5*) 0.2, and (HSF, *6*) 0.1.

Generalized parameter Θ, which indicates the fraction of polymer matrix for the formation of layers between particles in PFPCM, was calculated as follows:

$$\Theta = (\varphi_m - \varphi_f)/\varphi_m. \quad (2)$$

The composites of different compositions and structures were obtained using a Brabender plastograph with a chamber volume of 100 cm^3 at a temperature of 220°C; the rotor speed was 60 rpm, and the processing time was 10 min. The composition of the prepared PFPCMs based on LDPE + ShSO-30 was verified by burnout in a muffle furnace with subsequent determination of the filler fraction.

The rheological properties (viscosity) of LDPE + ShSO-30 dispersed systems with different compositions and structures were determined using an IIRT installation and an LCR7000 capillary rheometer (Dynisco, United States) at temperatures of 180, 200, 220, 240, and 280°C in wide ranges of velocities and shear stresses.

Figure 1 presents the dependences of the effective viscosity of PFPCMs with different structure types based on LDPE + ShSO-30 on the processing temperature at different values of generalized parameter Θ.

The viscosity of PFPCMs based on LDPE + ShSO-30 increases ~25 times compared with the LDPE viscosity at processing temperatures of 180 and 280°C when the filler is embedded and generalized parameter Θ is reduced from 1.0 to 0.10 vol pts. When the processing temperature is increased by 100°C for the same structure type (DS, LFS, MFS, or HFS), the viscosity increment of about six times can be eliminated, which is an effective parameter for regulating technological modes for molding products with different wall thicknesses. However, the time of thermal stability reduces as temperature increases and thermal oxidative destruction of polymers is observed.

The pressure loss during injection molding is observed with the growth of PFPCM viscosity, which limits the processing of polymer materials into products.

Classification of molding PFPCMs with various structure types (DS, LFS, MFS-1, MFS-2, and HFS) by fluidity was suggested in [12]: high-fluid (low-viscosity), fluid (viscous), and low-fluid (high-viscosity).

The classification of molding PFPCMs by processing properties, taking into account the dispersion structure, allows consideration of filling injection molds with melts from a new point of view. Thus, pressure loss ΔP_m can be connected not only with the viscos-

Table 1. Dimensions of plates for calculating pressure losses during injection molding

Plate parameters	Units	Value				
Length (L)	mm	100				
Breadth (B)	mm	50				
Thickness (h)	mm	0.5	1.0	3.0	5.0	8.0
L/h	–	200	100	33.3	20	12.5
Volume (V_{prod})	cm^3	2.5	5.0	15.0	25.0	40.0

Table 2. Specifications of injection molding machines produced by Bole Intelligent Machinery Co. (China)

Model	Ultimate characteristic		
	injection volume, cm^3	injection velocity, cm^3/s	molding pressure, MPa
BL70EKS (A)	55	60	320
BL230EKS (A)	315	140	280
BL470EKS (A)	1425	360	210
BL750EKS (A)	2510	545	230
BL1200EKS (A)	4670	820	215
BL1850EKS (A)	8195	1105	235
BL2800EKS (A)	23710	1700	210
BL4000EKS (A)	57750	2215	208

ity of dispersed systems, but also with types of structures of molding composite polymer materials, the compositions of which can be designed as shown in [13].

Pressure losses during filling injection molds at different temperatures with PFPCMs of various structure types (DS, LFS, MFS-1, MFS-2, and HFS) were calculated on the example of "plate"-type product with a length of 100 mm, a breadth of 50 mm, and wall thicknesses of 0.5 and 1.0 mm (thin-walled), 3.0 and 5.0 mm (average wall thickness), and 8.0 mm (thick-walled). The parameters of the samples are given in Table 1.

The viscosity of PFPCMs of various structure types depends on the viscosity of the initial polymer. Limitations connected with the processing of PFPCMs to products are determined by viscous properties, power-speed parameters of modern injection molding machines, and the developed maximum injection pressure (P_{inj}).

As an example, Table 2 lists the specifications of modern molding machines manufactured by Bole Intelligent Machinery Co. (China).

Molding machines with higher injection pressure are used to mold small-sized thin-walled products, while a high rate of melt injection is important for large-sized products.

Pressure losses in the injection molding machine and gating system were not considered in the calculation of pressure loss in the injection mold; melt temperature T_{m} was counted as an average flowing temperature of the melt $T_{\text{m}} \approx T_{\text{av}} = \text{const}$. The filling rate of the mold with PFPCM melt Q was 30 cm^3/s, and injection nominal volume V_{inj} was ~15% more than product volume V_{prod}.

Modern injection molding machines of the aforementioned series can provide the maximum injection pressure to 320 MPa (Table 2). Therefore, the injection pressure limit was $P_{\text{inj}} \leq 300$ MPa.

For steady flow conditions of PFPCM melts, pressure loss at injection mold filling ΔP_{m} was calculated by formula (3), taking into account the product thickness:

$$\Delta P_{\text{m}} = \eta Q_{\text{inj}} \frac{4L}{Bh^3}, \tag{3}$$

where η is the efficient viscosity of PFPCM melt at processing temperature, Pa s; Q_{inj} is the injection volume velocity, cm^3/s; L is the product length, cm; B is the product breadth, cm; and h is the product thickness, cm.

Figures 2–4 present the dependences of pressure loss at filling of the injection mold (ΔP_{m}) with PFPCM melts having different structure types (DS,

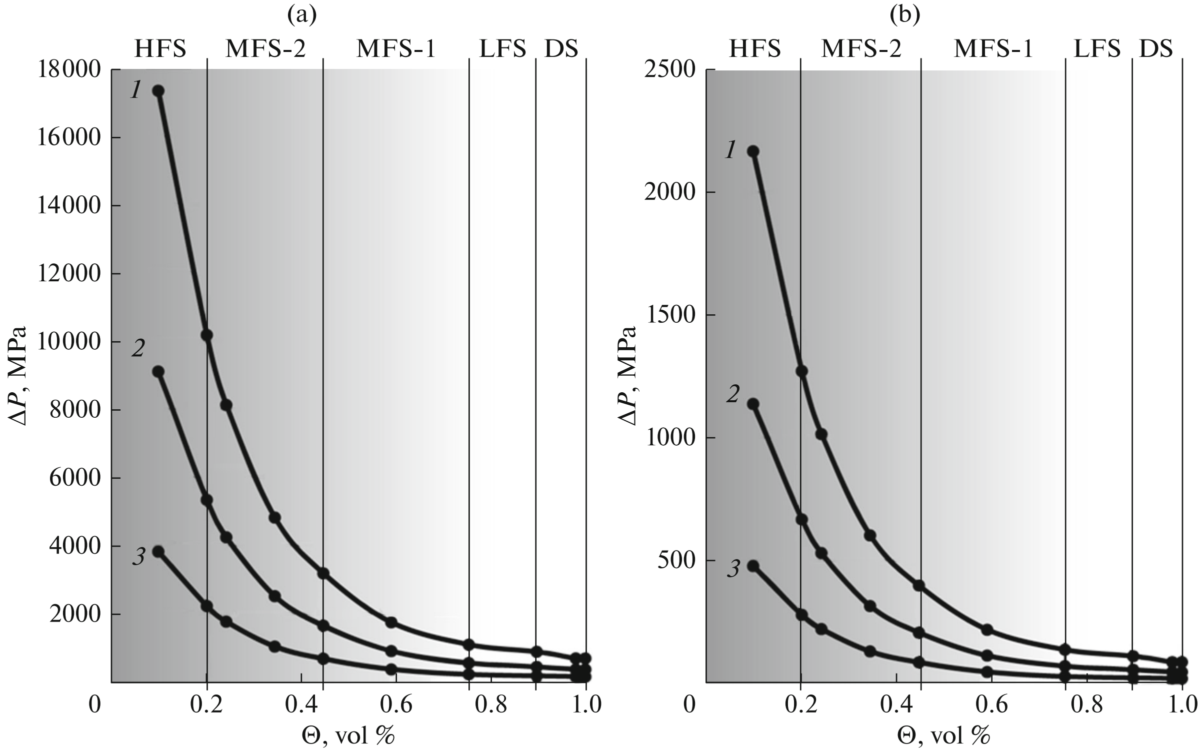

Fig. 2. Dependence of pressure loss during filling the injection mold with PFPCM on generalized structure parameter Θ for products with a thickness of (a) 0.5 and (b) 1.0 mm at different processing temperatures of (*1*) 180, (*2*) 220, and (*3*) 260°C.

LFS, MFS, and HFS) on generalized structure parameter Θ. The dependences were obtained for products of different thicknesses at various temperatures (processing temperature ranges).

The highest values of pressure losses (ΔP_m to ~4000–1730 MPa) were observed during filling the mold with PFPCM having highly-filled structure (HFS) with $0.2 \geq \Theta \rightarrow 0.0$ vol pts, product thickness of 0.5 mm, and injection molding temperature of 180–260°C (Fig. 2a). Such thin-walled products cannot be obtained by injection molding using MFS-2 and HFS PFPCMs.

According to the obtained results, thin-walled products (~0.5 mm) can be produced only using PFPCMs with DS, LFS, and MFS-1 types at a maximum melt temperature of 260°C, and pressure loss reaches ~300 MPa (ultimate parameters of injection molding machines are given in Table 2).

When the product thickness is increased to 1.0 mm, PFPCM of all structure types except HFS can be processed at 260°C.

When producing items with a thickness above 3 mm, manufacturing engineers have almost no complications using PFPCMs of all structure types (from DS to HFS) in a temperature range from 180 to 260°C. In this case, pressure loss does not exceed ~90 MPa, which is three times lower than the maximum pressure of injection molding machines (~320 MPa). However, pressure losses should be calculated, and the injection pressure capabilities of the injection molding machine should be taken into account when casting long products.

In the area of diluted systems (DSs) with $1.0 \geq \Theta \geq 0.90$ vol pts, pressure losses during injection mold filling with the PFPCM melt ($\Theta = 0.98$ vol pts and $\varphi_f = 0.01$ vol pts) are similar (the difference does not exceed 10%) to the LDPE injection molding in the whole temperature and thickness ranges.

Concerning low-filled systems (LFSs) with $0.9 \geq \Theta \geq 0.75$ vol pts, pressure losses during injection mold filling with the PFPCM melt are increased 1.1–1.5 times at 180 and 260°C compared with those of LDPE at similar conditions for the products of different thicknesses.

As for medium-filled systems (MFS-1 and MFS-2) with $0.75 \geq \Theta \geq 0.20$ vol pts, a pronounced growth of pressure losses (1.5–15 times) is observed in the whole temperature range of processing PFPCM products. In the case of MFS-1 (below the yield point) with $0.75 \geq \Theta \geq 0.45$ vol pts, pressure loss at filling the injection mold with PFPCM melt is increased ~1.5–5 times. When the yield point is reached and the PFPCM structure is MFS-2 ($0.45 \geq \Theta \geq 0.20$ vol pts), pressure losses increase ~5–15 times.

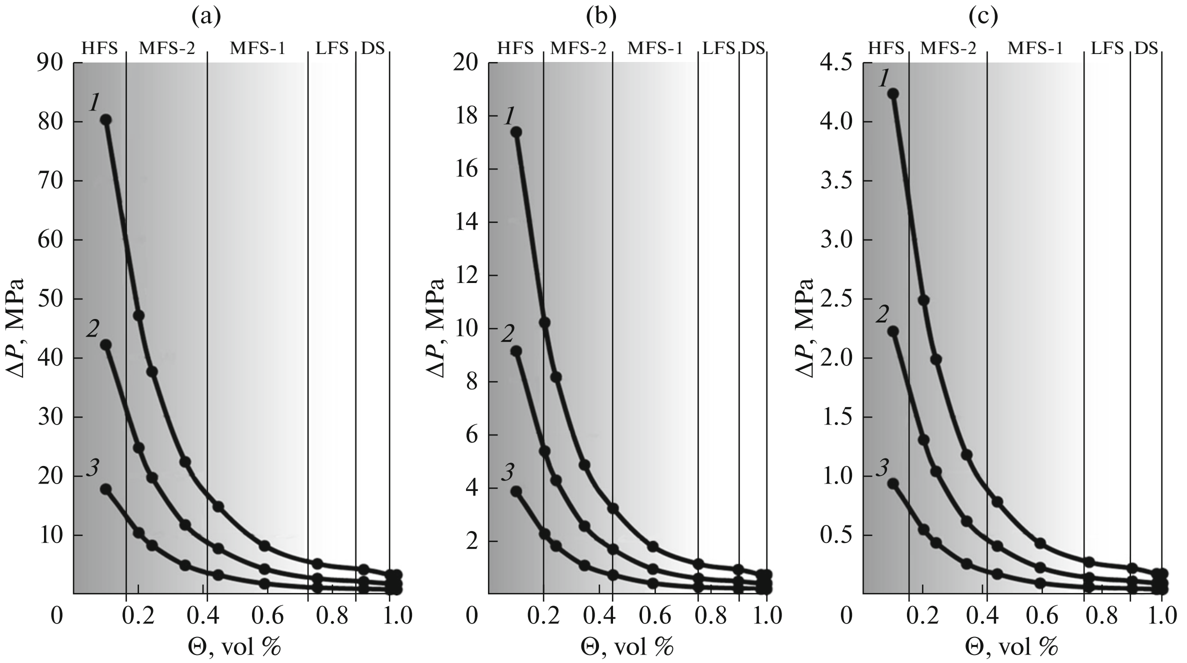

Fig. 3. Dependence of pressure losses during injection mold filling with PFPCM on generalized parameter Θ for a product with a thickness of (a) 3.0, (b) 5.0, and (c) 8.0 mm at different processing temperatures of (*1*) 180, (*2*) 220, and (*3*) 260°C.

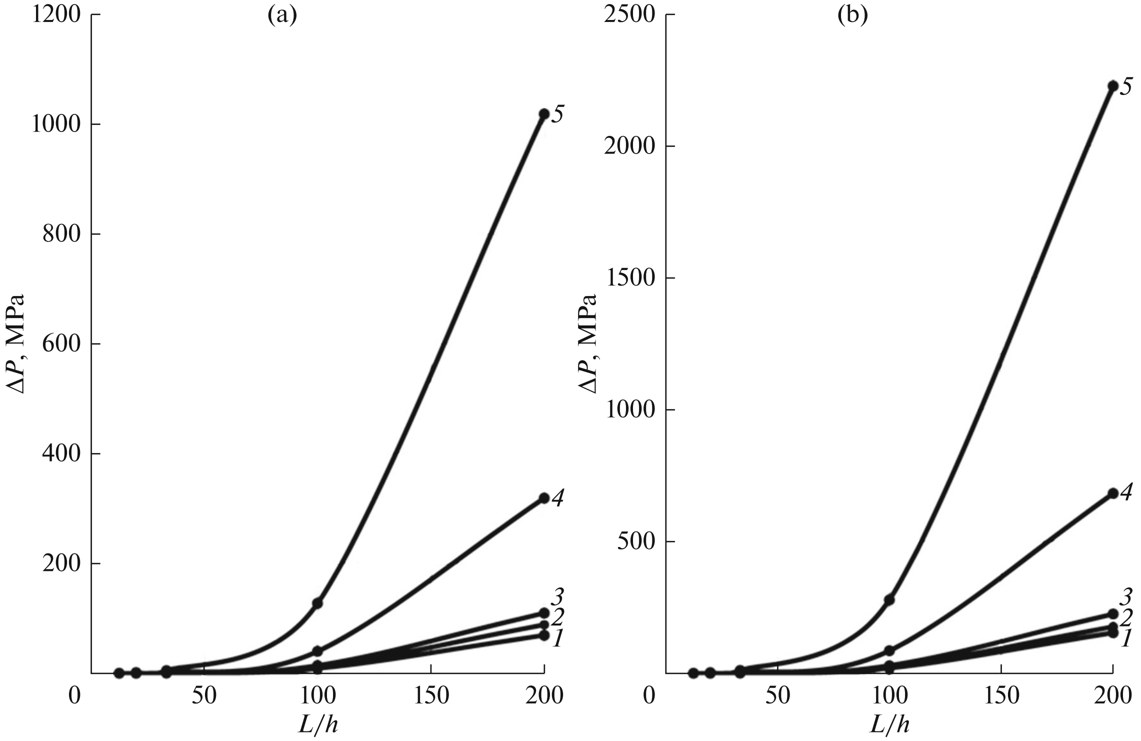

Fig. 4. Dependence of pressure losses in the injection mold on *L*/*h* parameter at processing temperatures of (a) 180 and (b) 260°C at different values of generalized parameter Θ: (DS, *1*) 0.98, (LFS, *2*) 0.90, (MFS-1, *3*) 0.75, (MFS-2, *4*) 0.50, and (HFS, *5*) 0.20.

In the case of highly filled (HFS) PFPCM with $0.20 \geq \Theta \to 0.00$ vol pts, pressure losses at mold filling reach the maximum: the increase is 15–25 times.

It is necessary to calculate the pressure loss at the use of PFPCM with various dispersion structures (DS, LFS, MFS-1, MFS-2, and HFS) and deliberately choose the brand of injection molding machine according to its passport data when adjusting the injection molding process for a product with a given thickness and length.

The reduced parameter—the ratio of length to thickness (L/h)—is used to estimate the effect of the product geometry on pressure losses at PFPCM injection molding. Figure 4 presents calculated dependences of pressure loss for PFPCMs with different structure types on the reduced parameter L/h at various temperatures.

An increase in temperature to 260°C makes it possible to obtain products with $L/h \approx 200$ using PFPCMs with structure types from DS to MFS-1.

According to the analysis of the dependences, obtaining products with $L/h \geq 50$ is complicated at low injection molding temperatures (180°C) for almost all types of PFPCM structures. When the injection molding temperature is increased to 260°C, items with $L/h \geq 100$ can be obtained using PFPCMs with DS, LFS, and MFS-1 structure types. However, when casting long products, pressure losses should be calculated and the injection pressure capabilities of the injection molding machine should be taken into account.

The dependences of pressure loss during filling the injection molds obtained for the first time are given in Figs. 2–4; these dependences are closely connected with the PFPCM structure (DS, LFS, MFS, and HFS).

Such an approach makes it possible to construct PFPCM compositions with various processability at the stage of the design of injection molding technology and intentionally choose the construction of an injection molding machine taking the product form factor into account.

For each product, similar calculations are carried out, pressure losses are determined and an injection molding machine is chosen for a PFPCM with a given structure type: DS, LFS, MFS-1, MFS-2, or HFS. According to the models of injection molding machines (IMMs) given in Table 2 and taking into account calculated values of pressure losses, sizes, and volumes of products, a BL70EKS (A) injection molding machine can be recommended.

Using a BL70EKS (A) injection molding machine, a molded item of the "plate" type with a thickness of 0.5 mm ($L/h = 200$) can be produced at a melt temperature of 240°C using PFPCMs based on LDPE + ShSO-30 having generalized parameter $\Theta \geq 0.90$ vol pts and DS structure type. When the melt temperature is 280°C, a product with $\Theta \geq 0.60$ vol pts can be obtained using composites with DS, LFS, and MFS-1 structures.

When the melt temperature is below 200°C, it is hardly possible to produce items with a thickness of 0.5 mm by injection molding because pressure losses are significantly higher than the maximum molding pressure ($P_{\text{inj. max}} \sim 300$ MPa) provided by a BL70EKS (A) machine.

An item of the "plate" type with a thickness of 1.0 mm can be obtained at a melt temperature of 180°C using PFPCMs based on LDPE + ShSO-30 having the generalized parameter $\Theta \geq 0.6$ vol pts (DS, LFS, and MFS-1). When the melt temperature is increased to 280°C, items with $\Theta \geq 0.2$ vol pts can be prepared (DS, LFS, MFS-1, and MFS-2).

Products with a thickness of 3.0, 5.0, and 8.0 mm can be molded using PFPCMs based on LDPE + ShSO-30 having the generalized parameter $1.0 \geq \Theta \geq 0.1$ vol pts and almost all structure types from DS to MFS-2 ($0.99 \geq \Theta/B \geq 0.20$) in the whole processing temperature range (from 180 to 280°C).

CONCLUSIONS

The choice of technological parameters and injection molding machine model depends on the item size (thickness, length, reduced parameter L/h), type of PFPCM structure, viscosity, and pressure losses during injection.

For the first time, the dependences of pressure losses during injection molding were established on generalized parameters, the type of dispersed PFPCM structure, and the dimensions of the products.

For thin-walled products with a thickness of less than ~1 mm, injection molding is complicated if PFPCMs with MFS-1, MFS-2, and HFS structure types are used.

It was shown that products with a wall thickness of more than ~3 mm and $L/h \leq 100$ can be obtained by injection molding using almost all PFPCM structure types (DS, LFS, MFS-1, MFS-2, and HFS) at processing temperatures. However, as L/h increases, difficulties may arise in injection molding processing.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

REFERENCES

1. A. E. Sorokin, V. A. Sagomonova, A. P. Petrova, and L. V. Solovyanchik, "Technologies for producing polymer composite materials based on a thermoplastic matrix (review)," Trudy VIAM, No. 3, 78–86 (2021).
2. M. N. Tukhtasheva, B. Zh. Saparov, and S. E. Normurodov, "Technological regimes for processing antistatic-heat-conducting, anti-friction, wear-resistant and

nanocomposite polymer materials," Universum: Tekh. Nauki, No. 2–2, 26–29 (2022).

3. S. P. Tuzhilin, Yu. A. Lopatina, and A. S. Sviridov, "Processing of polymer materials by free casting in vacuum," Vestn. Belgorod Gos. Tekhnol. Univ., No. 7, 93–100 (2020).
4. I. I. Fatoev, S. S. Ismatov, and B. B. Aslonov, "Strength and defectiveness of filled polymer materials," Universum: Tekh. Nauki, No. 4–4, 54–57 (2021).
5. A. S. Borodulin, A. N. Kalinnikova, S. S. Muzyka, and A. G. Tereshkov, "Polyetherimides for the creation of heat-resistant polymer composite materials with high physical and mechanical properties," Vestn. Belgorod Gos. Tekhnol. Univ., No. 11, 94–100 (2019).
6. K. I. Donetsky, D. V. Bystrikova, R. Yu. Karavaev, and P. N. Timoshkov, "Polymer composite materials for creating elements of aircraft transmissions (review)," Trudy VIAM, No. 3, 82–93 (2020).
7. E. L. Kalinchev and M. B. Sakovtseva, *Properties and Processing of Thermoplastics: Reference Manual* (Khimiya, Leningrad, 1983) [in Russian].
8. V. N. Matveenko and E. A. Kirsanov, "Viscosity and structure of disperse systems," Vestn. Mosk. Gos. Univ., Ser. 2 Khim., No. 4, 243–276 (2011).
9. V. N. Matveenko and E. A. Kirsanov, "Structural justification of non-Newtonian flow," Vestn. Mosk. Gos. Univ., Ser. 2 Khim., No. 2, 59–81 (2017).
10. D. D. Krechetov, A. N. Kovaleva, and I. D. Simonov-Emelyanov, "Rheological properties of dispersed-filled thermoplastics with different types of structures at processing temperatures," Plast. Massy, Nos. 9–10, 19–22 (2020).
11. I. D. Simonov-Emelyanov, K. I. Kharlamova, and E. R. Dergunova, "Oil absorption capacity of dispersed powders and determination of the maximum content of fillers in polymer composite materials," Klei. Germetiki. Tekhnol., No. 3, 18–24 (2022).
12. D. D. Krechetov and I. D. Simonov-Emelyanov, "Structure, generalized parameters and rheological properties of dispersed-filled thermoplastics," Materialovedenie, No. 9, 38–44 (2020).
13. I. D. Simonov-Emelyanov, D. D. Krechetov, and K. I. Kharlamova, Design of compositions and types of structures of injection molded dispersed-filled thermoplastics with good processability and high strength," Plast. Massy, Nos. 5–6, 10–12 (2021).

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