

FORMATION OF FIBER MATERIALS BY PNEUMATIC SPRAYING OF POLYMERS IN VISCOUS-FLOW STATES

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Using a novel ejection spraying unit and relying on new approaches, fibers are formed by the method of pneumatic melt blowing of polycarbonate, polypropylene, and polyethylene terephthalate. The proposed approach is based on the concepts of atomization of the polymer melt flow as a preferential regime for fiber-material formation. From the analysis of the values of numerical characteristics in the zone of atomization and the physical background of the criteria under study a conclusion is drawn that the essential role in destruction of the jet belongs to the formation of a boundary layer in the melt under the action of friction forces, followed by its separation. An assumption is made on the prevailing action of the separating destruction of the melt jet via the mechanism of 'skinning' of the boundary layer of the melt due to a shorter time of its persistence compared to the development of the Kelvin–Helmholtz instability.

Keywords: fiber material, fiber formation, method of pneumatic spraying.

INTRODUCTION

The advances in understanding the processes taking place during the formation of synthetic fibers favor an extension of the product line made from them and hence their increasingly wider general application [1]. Conventionally, these processes rely on extrusion of the viscous-flow polymers through the dies. A considerably high viscosity and low elasticity of the melts make the polymer flow through the dies difficult. In this connection, new approaches are sought for in order to develop fiber-formation processes. A special attention is given to a promising technology of direct aerodynamic formation of fiber materials from the melt, which offers a possibility of designing materials with special functional characteristics [2, 3]. Of particular promise are the methods of fiber formation under direct impact of an actuating medium (gas, vapor, aerosols) on a freely flowing jet of the polymer melt formed using ejectors. In these devices, the fiber-forming polymer melt is sprayed due to the energy of the gas flow generally forced at near sonic velocities, followed by its solidification as fibers. The processes for pneumatic spraying of a freely flowing jet due to their technological features are most suitable for spraying of contaminated and inhomogeneous melts [4]. On the other hand, there are no sufficient physical concepts for optimization of the rational spraying regimes. In view of this fact, attempts are made to use the fundamental principles of hydraulics and gas dynamics with a purpose of analyzing the process of dispersion and formation of a spray pattern.

Thus, the purpose of this work is to perform investigation of the influence of the processes taking place during spraying of a freely outflowing jet of the melt on the size distribution of the resulting polymer fibers.

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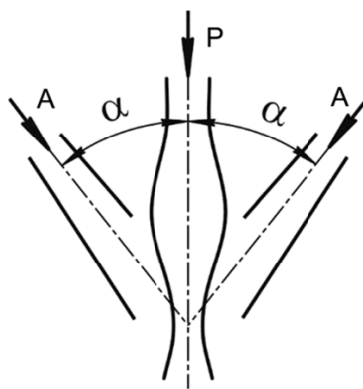


Fig. 1. Schematics of the air flow impact (A) onto the jet of the dispersed polymer melt (P).

1. MATERIAL, EXPERIMENTAL PROCEDURE, APPROXIMATIONS

The specimens of fiber material (FM) were formed from the melt of thermoplastic polymers having different physical and rheological properties. The initial materials were: Makrolon 2456 polycarbonate (PC) (Bayer, Germany), PP H080 GP polypropylene (PP) manufactured in accordance with TU 2211-103-70353562-2013, abr. 3 (Russia), and scrap polyethylene terephthalate product (SPET) in the form of flakes. The rheological characteristics of the SPET melt were determined using an IIRT-AM extrusion rheometer in accordance with the GOST 11645-73 requirements. The melt density was determined according to the DIN 53735 standard by the Verfahren B method, and the surface tension was determined using a JYW-200C tensiometer (du Noüy type) according to the GOST 20216-74. The characteristics of commercially produced PP and PC melts were determined in [5].

The dimensions of FMs were made by the method of dispersion analysis using the data of a microscopic examination. The microimages of FMs from PP (PPFMs) and SPET (PETFMs) were obtained using a Hitachi TM-1000 scanning electron microscope at the accelerating voltage 15 kV. The microscopic examinations of the FM specimens from PC (PCFMs) were performed using an Olympus GX-71 optical microscope.

Fiber materials were formed using a laboratory setup by the method of pneumatic spraying of a freely outflowing melt jet. The key element of the setup is an air-ejection device with a unique design of the annular convergent nozzle. The polymer specimens were transformed into a viscous-flow state by their melting in a laboratory reactor equipped with a stirring device. The melt temperature is controlled by a TRM1 measuring-regulating device with a chromel-alumel thermocouple. The melt from the reactor was fed into the fiber-forming unit via a cylindrical channel within which a continuous vertical jet of the melt was dispersed by the compressed-air flow. The resulting fiber material was precipitated onto a mesh transporter on the bottom of the vertical chamber.

Clearly, of great importance is the scheme of the impact of the actuating medium onto the jet of the sprayed polymer melt, therefore note should be made of the nozzle design features. We realized a concurrent scheme of dispersion at the angle $2\alpha = 20^\circ$ (Fig. 1), which allowed forming necessary ejection flows. The ejection flow involves the melt jet into the dispersion zone, preventing the contact between the melt and the dispersing unit surface, and ensures a possibility of forming functional coatings on the fibers during their formation, introducing aerosols into the ejection contour.

Since fiber formation occurs due to the disintegration of a jet of the melt into thin fibers during polymer transition from the viscous-flow into the solid state before the onset of a repeated decay and also due to the absence of a contact between the melt and the parts of the dispersing unit, the energy carrier selected was air at $T_A = 20^\circ$.

It is well known that stable dispersion begins at the ratios of the gas-to-fluid flow kinetic energies higher than two [6]. Moreover, it could be assumed that during the initial jet disintegration the melt viscosity is low and does not

substantially affect its melt blowing. Also, the melts are thought to be pseudoplastic non-Newtonian fluids and the process of their dispersion is characterized by high velocities.

If the ratio of the kinetic energies of the air and polymer flows is

$$\frac{\rho_a \cdot V_a^2}{\rho_p \cdot V_p^2} > 2, \quad (1)$$

then the velocity ratio would be

$$\frac{V_a}{V_p} > \sqrt{2 \frac{\rho_p}{\rho_a}}. \quad (2)$$

Here ρ_a and ρ_p are the air and polymer melt densities and V_a and V_p are the air and melt flow velocities. It follows from (2) that in the case of dispersion of the PP melt ($\rho_p = 739 \text{ kg/m}^3$) by air ($\rho_a = 1.205 \text{ kg/m}^3$) the air flow velocity would be at least 35 times that of the melt flow velocity, and in the case of dispersion of SPET ($\rho_p = 1100 \text{ kg/m}^3$) by air – 43 times higher than the melt flow velocity.

The average gas flow velocities in the characteristic cross-sections of the ejection spray gun were found using a specially designed application, EJFLUENT [7], whose computational algorithm is based on the equations of gas dynamics for the air flow rate Q

$$Q = \mu A \sqrt{2 \frac{\Delta P}{\rho}}, \quad (3)$$

where μ is the flow rate coefficient found experimentally, A is the cross-section area of the opening from which the melt flows, ΔP is the pressure difference causing flow, and ρ is the gas density. Then the average effluent gas flow velocity V would be

$$V = \frac{Q}{A}. \quad (4)$$

The flow regime was determined from a comparison of the pressure difference ($\beta = P/P_0$) with the critical ratio

$$\beta_{cr} = \frac{P_{cr}}{P_0} = \left(\frac{2}{k+1} \right)^{\frac{k}{k-1}}, \quad (5)$$

where P and P_{cr} are the pressure values in the input and critical cross-sections, P_0 is the input nozzle pressure, and k is the adiabatic exponent. Note that the critical pressure ratio does not depend on the deceleration parameters but is a function of the physical gas properties only. The carrier gas was air for which $k = 1.4$, so $\beta_{cr} = 0.528$.

Under the subsonic regime, the average velocity of air flow from an annular convergent nozzle was determined by the following expression [8]:

$$V = \sqrt{\frac{2k}{k-1} R (T_0 - T)}, \quad (6)$$

and under the supersonic regime, the average velocity was assumed to be equal to the sound velocity

TABLE 1. Characteristics of Initial Melt Substances for Synthesis of Fiber Materials

Physical properties	PP	SPET	PC
Density, kg/m ³	910	1350	1200
Melt temperature, °C	250	230	300
Melt flow index, g/ 10 min	8	18	20
Melt density, kg/m ³	739	1100	1050
Melt viscosity at low shear velocity, Pa·s	582*	384*	275
Melt surface tension, mJ/m ²	13.1	29.8	24.6
Melt mass flow rate, kg/h	16.2 ± 1.1	25.2 ± 1.8	21.6 ± 1.6
Melt jet velocity upon reaching the dispersion zone, m/s	1.56	1.58	1.55

*Calculated according to [9].

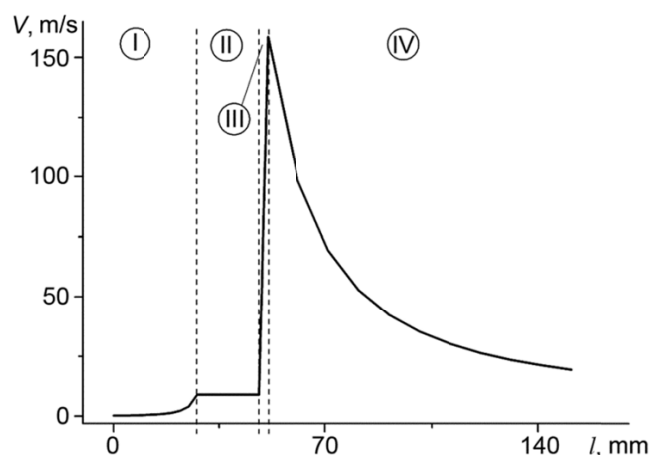


Fig. 2. Variation of the average air-flow velocity along the fiber-forming setup.

$$a = \sqrt{kRT} , \quad (7)$$

where R is the universal gas constant and T_0 and T are the nozzle inlet and outlet air temperatures, respectively.

There were some other assumptions. The velocity distribution was taken to be uniform for every cross-section under investigation. Gas compressibility was not taken into consideration, and all thermodynamic gas parameters were thought to be constant during the entire process. It was also assumed that the velocity value in the annular nozzle outlet place obeyed the linear law.

2. RESULTS AND DISCUSSION

For the estimations we used the physical and rheological characteristics of polymers [5] and the values of mass flow rate and velocity during dispersion (controlled during the experiment), which are given in Table 1.

The variation in the average air flow velocity along the axis of the fiber-forming setup is shown in Fig. 2. The estimates were made using (3)–(7); the dashed line indicate the boundaries of its characteristic sections.

It is clear that the highest value of the cross-section-average velocity is achieved in Section III, where the nozzle outlet is located (Fig. 2). The melt dispersion is, however, realized in the cross-section where the air flows converge and the average velocity is found to be $V_{av} = 52.8$ m/s. It is logical to assume that the distribution of the

absolute value of velocity would be a Gaussian, and the velocity would be close to zero near the walls. Given this, we can argue that the maximum velocity would be observed in the region where the flows converge, where it will be $V_{\max} \approx 100$ m/s. Therefore, the values of the air flow-to-polymer melt flow velocity ratio V_A/V_P would lie within the interval from approximately 63.3 to 64.5, which is a sufficient condition for formation of a steady dispersion jet of all polymers under consideration.

In fiber-material formation, a significant role belongs to the regime of the polymer-melt disintegration [6]. The most important role among the governing parameters affecting the disintegration regime is thought to belong to the Weber number We_j for a jet, which characterizes the ratio of the inertia forces to the surface tension forces

$$We_j = \frac{\rho_a (V_a - V_p)^2 d_p}{\sigma}. \quad (8)$$

Here d_p is the polymer jet diameter determined from the mass balance equation (4) and σ is the melt surface tension. A considerably large value of We_j indicates that jet disintegration can follow two mechanisms. The first is separation of micro drops from the wave crests on the upstream face of the jet, and the second mechanism is its catastrophic disintegration.

The Reynolds criterion Re_j for a jet characterizes the ratio of the inertia forces to the friction (viscosity) forces

$$Re_j = \frac{\rho_a (V_a - V_p) d_p}{\mu_a}. \quad (9)$$

A considerably large value of Re_j suggests a turbulent regime of air flow.

The Laplace criterion Lp_j characterizes the ratio of the surface tension forces to the friction (viscosity) forces

$$Lp_j = \frac{\rho_p d_p \sigma}{\mu_p^2}. \quad (10)$$

At small values of Lp_j , a transition from the skinning disintegration, taking place according to the mechanism of stripping the melt boundary layer, to the mechanism of the Kelvin–Helmholtz instability development is impossible

$$We \geq 1.65 \cdot 10^4 \cdot Oh^{0.4}, \quad (11)$$

where $Oh = Lp^{-0.5}$ is the Ohnesorge number.

The generalized criterion [5] An_j for a jet includes the Weber We_j , Reynolds Re_j and Laplace Lp_j criteria

$$An_j = d_p \sqrt{\frac{\rho_a^3 (V_a - V_p)^3}{\sigma \rho_p \mu_p}}. \quad (12)$$

The calculated numerical values of the main criteria controlling the regimes of the jet disintegration in the dispersion zone during the fiber-material formation are presented in Table 2.

Relying on the analysis of the values of numerical characteristics in the dispersion zone and the physics behind these criteria, a conclusion can be drawn that the dominant role in the jet disintegration belongs to the formation of a boundary layer in the melt under the action of friction forces and its subsequent separation.

In the course of the experimental testing of the proposed approaches of fiber-forming materials, the relative air-flow and melt-jet velocities remained constant and so did the jet diameter. The polymer melt density, surface tension, viscosity, and temperature were varied (see Table 1). In all of the experiments, a stable dispersion of the polymer melts

TABLE 2. Values of Main Criteria in Dispersion Zone

Similarity criteria	PP	SPET	PC
We_j	1990.2	890.6	1045.6
Re_j	$15.1 \cdot 10^3$	$15.4 \cdot 10^3$	$14.9 \cdot 10^3$
Lp_j	$6.37 \cdot 10^{-8}$	$5.1 \cdot 10^{-7}$	$7.5 \cdot 10^{-7}$
An_j	$1.2 \cdot 10^{-3}$	$0.83 \cdot 10^{-3}$	$1.07 \cdot 10^{-3}$

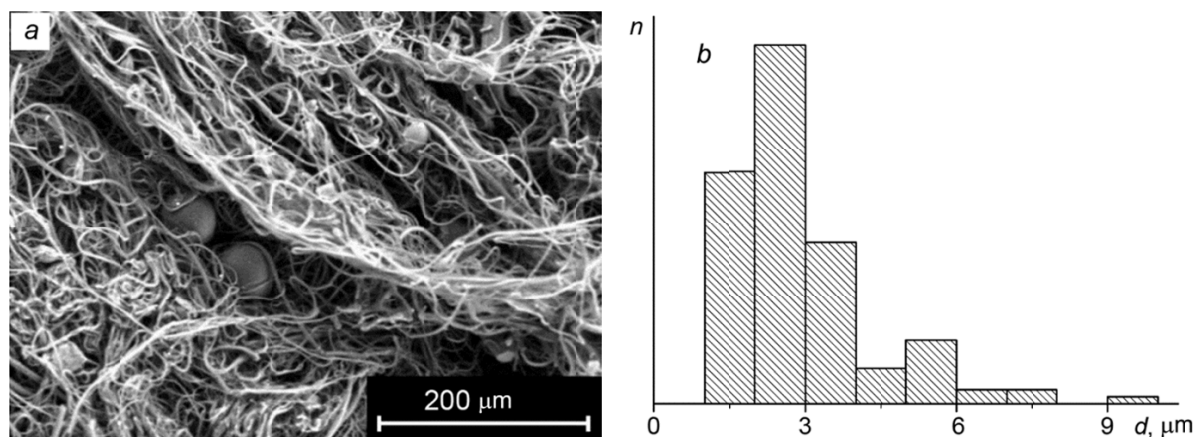


Fig. 3. SEM image of PPFM specimen (a) and bar chart of fiber diameter distribution (b).

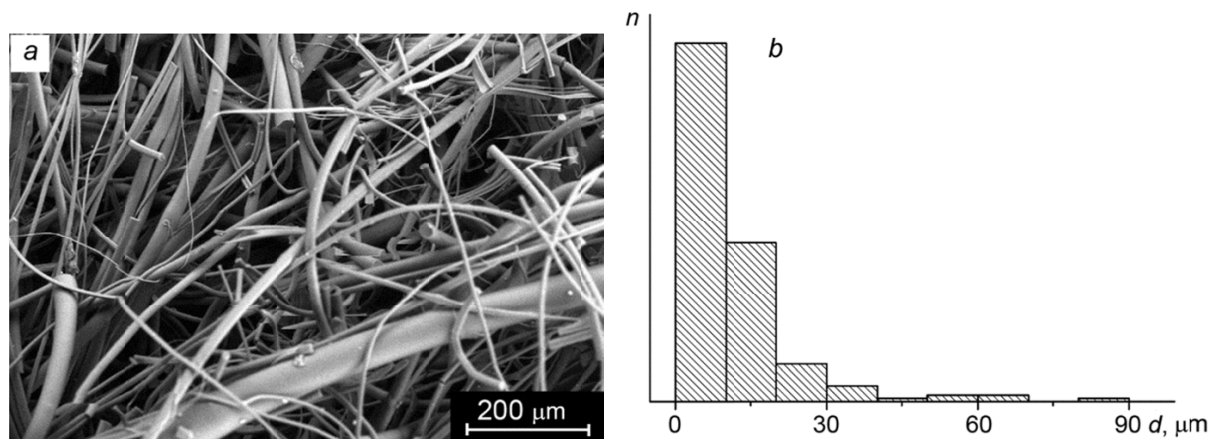


Fig. 4. SEM image of SPETFM specimen (a) and bar chart of fiber diameter distribution (b).

was observed, which was followed by the fiber formation. Figures 3a and 4a present the images of PPFM and SPETFM specimens, respectively, obtained using scanning electron microscopy (SEM) and Fig. 5a shows the image of a PCFM specimen obtained using optical microscopy (OM). We failed to obtain SEM images of PCFM specimens due to the effect of secondary electron emission. Figures 3b, 4b, and 5b present the histograms of fiber diameter distribution.

The statistical characteristics of the fiber size distribution presented in Table 3 suggest that all histograms are characterized by positive symmetry. The curves from the PPFM and SPETFM specimens have higher peaks (leptokurtic distribution) compared to those of a normal distribution. The high value of the excess kurtosis of the diameter distribution curve from the SPETFM specimens implies a large number of fibers with hardly varying diameters. The

TABLE 3. Statistical Characteristics of Fiber Size Distribution

Specimen	Average diameter \bar{d} , μm	Standard deviation σ	Median	Skewness	Excess kurtosis
PPFM	3 ± 0.25	1.47	2.94	1.81	1.02
SPETFM	12.45 ± 1.68	11.67	9.60	3.06	9.64
PCFM	57 ± 7.48	29.68	55.77	1.22	-1.03

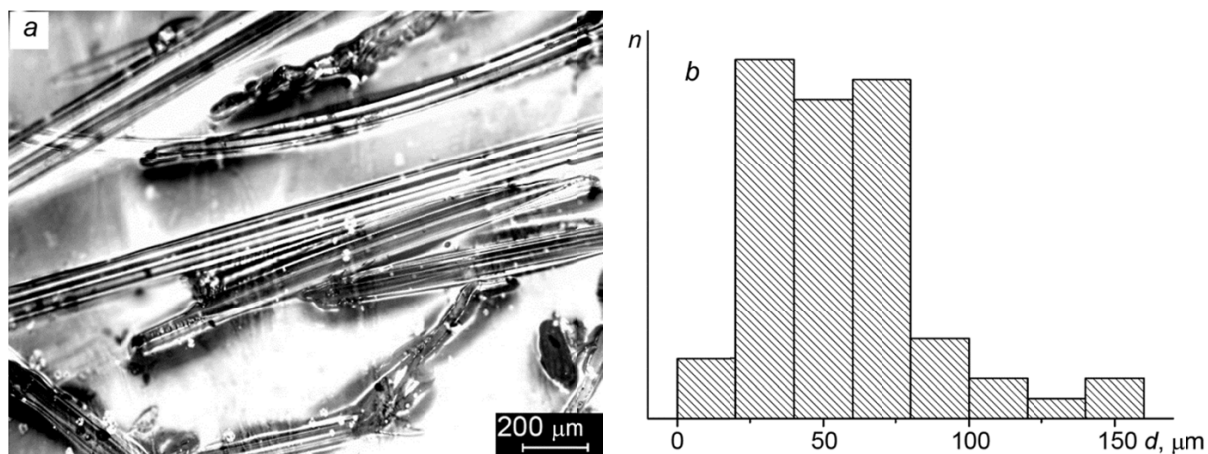


Fig. 5. OM image of PCFM specimen (a) and bar chart of fiber diameter distribution (b).

negative value of the excess kurtosis of the diameter distribution curve from the PCFM specimens suggests the presence of a large number of fibers with the near-average diameters. In addition, the PPFM and SPETFM specimens contain a small amount, no more than 10%, of large fibers. The PPFM specimens contain a small number of non-fibrous spherical inclusions with the average diameter about 40 μm . From the presence of a large number of the fiber end sections in the images of the SPETFM and PCFM specimens one can also judge about brittleness of the resulting fiber materials.

SUMMARY

Using a novel ejection-type dispersion setup and relying on a new approach, fibers have been formed from the melts of polycarbonate, polypropylene, and polyethylene terephthalate by the method of pneumatic spraying. The proposed approach relies on the concepts of the primary importance of the regimes of the polymer melt jet disintegration for the fiber material formation.

A conclusion has been made, based on the analysis of the numerical characteristics in the dispersion zone and relying on the physical meaning of these criteria, that the leading role in the jet disintegration belongs to the formation of a boundary layer in the melt under the action of friction forces and its subsequent breakaway. An assumption has been made on the prevailing influence of the breakaway jet disintegration via the boundary-layer skinning due to a shorter time of its persistence compared to the Kelvin – Helmholtz instability development.

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