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DETERMINING THE EFFECT OF DISPERSED ALUMINUM PARTICLES ON THE FUNCTIONAL PROPERTIES OF POLYMERIC COMPOSITES BASED ON POLYVINYLIDENE FLUORIDE

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Polymeric materials that contain inorganic fillers demonstrate a unique set of physical properties due to the combination of matrix elasticity and filler strength. This paper reports determining the effect of dispersed aluminum particles on the properties of polyvinylidene fluoride-based materials. This study result is the fabrication of a series of composite materials using a piston extruder. Their functional characteristics have been explored using the methods of thermophysical and mechanical analysis, dilatometry, and acoustic spectroscopy. It was established that the introduction of dispersed aluminum particles leads to the loosening of the matrix, which may indicate the transition of macro macromolecules from the crystalline phase to the boundary layer around the filler. This feature of structure formation and the uniform distribution of filler particles ensured the improvement of the functional characteristics of the materials obtained. It has been shown that with an increase in the content of filler in the system to 5 % the thermal conductivity increases from 0.17 W/(m·K) to 1.55 W/(m·K). The introduction of the filler leads to an improvement in the heat resistance of the materials obtained, by 17 K. The increase in both melting point and destructiveness is explained by the formation of a more perfect polymer structure with a higher degree of crystallinity. An increase in the speed of ultrasound propagation was identified, by 67 %, as well as in the tensile strength, by 36 %, in the materials obtained, which can be explained by contributions from the filler, which has greater sound conductivity and mechanical strength than the polymer matrix. Such systems show the reinforcing effect of aluminum particles on the polymer matrix, so they could be used as structural materials with improved functional characteristics

Keywords: polymeric composite materials, dispersed aluminum particles, thermal conductivity, tensile strength, sound speed

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1. Introduction

Polymer composite materials (PCM) have been the subject of intensive research over the past two decades [1]. The peculiarity of this type of material is the presence in their composition of inorganic fillers of very small size, which, when interacting with a polymer matrix, improve its physical properties. Due to this, PCS have a very wide scope of application: from structural materials to coatings to sensors [2].

The decisive effect of fillers on the polymer matrix is ensured by the presence of a very large interaction surface. However, given that the particles are very small in size, there are very powerful van der Waals forces of attraction among them. This leads to a strong sticking of nanoparticles, that is, their aggregation [3]. The main issue related to polymer nanocomposite manufacturing is to overcome random aggregation to ensure uniform distribution of nanoparticles in the polymer matrix [4], or their directional structuring [5]. The main efforts of scientists are aimed at developing new approaches to the manufacture of PCM with uniform distribution of filler particles. Another trend associated with the manufacture of PCM is to make them cheaper. This approach is implemented by selecting a preparation technique and a relatively cheap filler to obtain PCM with functional characteristics on a level with more expensive analogs.

Therefore, research aimed at improving the functional characteristics of PCM and expanding the scope of their application is relevant. Promising in this direction is to combine a unique method of mixing PCM using a cheap filler, for example, dispersed aluminum particles, which could make it possible to obtain materials with improved functional properties.

2. Literature review and problem statement

It is stated in [6] that the decisive effect of fillers on the polymer matrix is ensured by the presence of a very large surface. However, given that the particles are very small in size, powerful van der Waals forces of attraction operate between them. This leads to a strong sticking of nanoparticles, that is, their aggregation [3]. To destroy such aggregates from fine particles, it is necessary to provide the system with very large energy. The main issue of polymer composite production is to overcome aggregation and ensure the uniform distribution of particles in the polymer matrix [7].

Various methods are used to achieve the uniform distribution of fine filler in a polymer matrix [8–12]. These methods have both their advantages and disadvantages. For example, mixing dry powders [8] does not ensure the destruction of the primary aggregates of the filler. Mixing in solution [9] ensures uniform distribution of the filler but cannot be used for most polymers that are almost insoluble. Mixing in melt [10] using a screw extruder cannot be used for thermoreactive polymers. *In situ* polymerization [11] and the sol-gel process [12] are chemically complex methods that require the selection of optimal synthesis conditions.

One of the most common methods used industrially is the polymer melt mixing method. This technique implies dispersing fine particles into a polymer matrix that is in a molten state (a viscous liquid state). The method of mixing in the melt was used by different researchers to create composites based on various polymer matrices and fillers, for example, silicon polypropylene-dioxide [13], polypropylene-nanoclay [14]. Work [15] reported composites based on polypropylene and montmorillonite by mixing in the melt with a two-screw mini-extruder at a temperature of 180 °C and a rotational speed of 300 rpm. Such high rotational speed and high shear deformations could not ensure complete exfoliation and uniform distribution of clay plates in the polymer matrix.

Thermoplastic polymers are used to mix with the filler in the melt. Given the acceptable set of physicochemical properties, poly(vinylidene fluoride) (PVDF) is a promising material to obtain PCM [16]. The PVDF matrix, due to its high heat resistance and reduced melt viscosity, could be easily processed with an extruder. Materials created on the basis of PVDF and fine particles of metals or their oxides, such as aluminum, are widely used. Thus, composite materials are made of PVDF fibers and Al_2O_3 nanoparticles. These membranes exhibit high thermal stability and electrochemical properties as separators for use in lithium-ion batteries [17]. Other authors investigated the thermal conductivity of PVDF-Al systems made using a double-shaft mixing mill [18]. It was established that with a high content of the filler, Al particles begin to form clusters and a more compact packaging structure inside the matrix. At a content of 60 % Al, the thermal conductivity of the composite reached approximately 1.74 W/(m·K), which is almost ten times higher than that of a non-filled PVDF. However, high thermal conductivity values were obtained at a very high filler content. The formation of large clusters of aluminum particles indicates an imperfection of the mixing system. At present, modern extruding methods (screw extruders and mills) are not always able to ensure uniform distribution of filler in the polymer matrix because they do not make it possible to control the mixing time in a wide interval.

Thus, current studies that tackle the structure and properties of composite systems based on polyvinylidene fluoride and dispersed metal particles do not answer many questions related to the creation of composites with improved characteristics. Research into the influence of microscale filler on the functional properties of polymer systems when applying new methods of manufacturing such composite materials requires further development.

3. The aim and objectives of the study

The aim of this work is to determine the effect of finely dispersed aluminum particles on the functional properties of polymer composite materials based on polyvinylidene fluoride, made with a piston extruder. This would make it possible to identify patterns of influence of filler on the final properties of the system and obtain polymer composite materials with improved functional characteristics.

To accomplish the aim, the following tasks have been set:

– to define the optimal conditions for the manufacture of composite materials and monitor the effect of the filler on the density of the materials obtained;

– to determine the effect of the filler on the thermal conductivity of manufactured materials;

– to investigate the influence of the filler on the thermal physical characteristics of the materials obtained;

– to establish the features of the influence of the filler on the physical and mechanical characteristics of the materials obtained.

4. Materials and methods to study the influence of filler on functional properties of polymer composite materials

4. 1. The studied materials used in the experiment

Polyvinylidene fluoride (M_w ~180,000) was manufactured by Fluka (Germany). The melting point is between 175–180 °C; the density at 25 °C is 1.78 g/cm^3. PVDF is non-toxic, resistant to heat and chemicals, has low water absorption characteristics.

Dispersed aluminum particles (Al) manufactured by Ferrotrad LLC (Ukraine) were used as a filler. Dispersed aluminum particles have a lamellar shape and are covered with a thin oxide film. The distribution of particles in size is 0.5–10 μm. The average linear particle size is 1 μm. The filler density is 3.9–4.0 g/cm^3, the specific surface is 150 m^2/g, the thermal conductivity coefficient is 29–40 W/m·K.

4. 2. The procedure of manufacturing the examined materials

Composite samples were manufactured by extruding (mechanical grinding in the melt) with a piston extruder, followed by cooling according to the room temperature. The content of the filler varied within (1÷5) % by weight (hereinafter – %). The main advantage of piston extruders over screw extruders is the ability to vary the time of mixing filler particles with molten polymer matrix, after which one could make a sample to be examined in the form of either a plate or a thread.

The manufacturing technology of the materials under investigation is given below. After preliminary preparation of components of the composite material using a method of mechanical mixing, the mixture was loaded into the extruder (Fig. 1). The mold was heated to 200 °C. The rotation of the piston ensures the mixing of the melt of the polymer composite due to the polymer's own relatively high viscosity.

Experimentally, it was established that the most uniform mixing of filler particles in the matrix occurs within 10 minutes. With a further increase in the mixing time, partial polymer destruction occurs. After 10 minutes, a hole was opened in the lower part of the mold and, without turning off the rotation of the piston, the pressure of the translational motion gradually increased. The shear stresses that arose caused the polymer composite to melt through the hole (diameter ~2 mm). After that, the resulting material in a filamentous form was placed in a mold for hot pressing, where we set the appropriate shape of the sample under study.

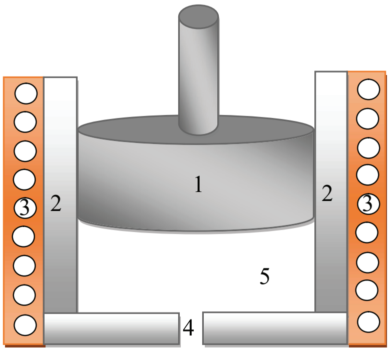

Fig. 1. Schematic representation of piston extruder: 1 — rotating piston; 2 — cylindrical base; 3 — heater; 4 — a hole in the base of the cylindrical mold; 5 — powdered composite

4. 3. Methods for studying the functional properties of obtained composite materials

The thermal conductivity of the samples studied was measured by the method of dynamic calorimetry using the device IT-λ-400 (Kazakhstan) (a thermal conductivity meter), with an improved cell [19]. The meter was calibrated by measuring the thermal conductivity of reference samples of quartz and copper. Measurements were carried out under a monotonous heating mode. To improve the accuracy of the measurement, the thermal conductivity of each sample was measured three times, followed by averaging the results. The measurement error was 3 %.

Thermal physical examinations were carried out in a dry air atmosphere with the help of an upgraded Perkin Elmer DSC-2 device (Germany). The temperature range was from 410 K to 560 K, the heating speed was 2 K/min, the research method is modulated DSC.

The speed of spread and the coefficient of ultrasonication were determined by the echo-pulse method using the US-12IM meter (Moldova). The ultrasound frequency was 5 MHz. To improve the accuracy, we measured each sample three times, followed by averaging the results. The measurement error was no more than 5 %.

The density of the studied composite materials was determined using the method of volumetric dilatometry, which is based on the method of hydrostatic weighing. Quartz and ethyl alcohol were chosen as references. The measurement error was no more than 2 %.

In this work, we studied destructive stresses at stretching according to (tensile strength). Dimensions of the samples studied: length, l=100±2 mm; diameter, d=2±0.3 mm. The study of the influence of filler contents on tensile strength was carried out using the automated breaking machine UM-5 (Ukraine) (modified with high-precision pressure and displacement sensors) at load speed υ=5 N/s. The measurement error did not exceed 3 %.

5. Results of studying the influence of filler on the functional properties of polyvinylidene fluoride-based materials

5. 1. Influence of fillers on the density of polyvinylidene fluoride-based systems

Everybody knows that the introduction of inorganic fillers significantly modifies the structure of the polymer matrix while changing the redistribution of crystalline and amorphous phases. As a result of such redistribution, the density of the composite material changes. Therefore, to determine the effect of the filler on the polymer matrix, we studied the dependence of the density of PVDF-based systems on the concentration of filler.

Fig. 2 shows the experimental results from determining a density value for the PVDF-Al systems. It is characteristic that at T=293 K the studied system demonstrates a nonlinear increase in the value of ρ when changing the contents of the filler in the range of (0÷5) %. At the same time, with an increase in the content of Al particles, the density of the materials obtained decreases.

Based on the law of additivity of the masses of polymer composite components, we calculated a value of the theoretical density ρ_T of the composite [20]:

$$\rho_T=\rho_M+\frac{\varphi_F}{\varphi_F+\frac{\rho_F}{\rho_M}\left(1-\varphi_F\right)}\left(\rho_F-\rho_M\right), \tag{1}$$

where ρ_M, ρ_F are the polymer matrix and filler density, respectively; φ_F is the mass content of filler.

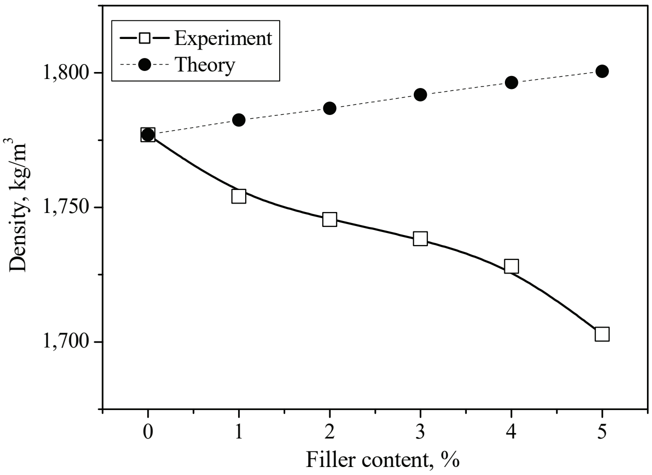

Fig. 2. Dependence of density on the content of dispersed aluminum particles for systems based on polyvinylidene fluoride

The experimentally obtained values for the density of PVDF-Al systems (Fig. 2), in accordance with [20], allow us to determine the value of composite defects in the form of porosity P:

$$P=1-\frac{\rho-\left[\frac{\rho_F}{\rho_M}\left(1-\varphi_F\right)\right]\rho_M}{\left[1-\frac{\rho_F}{\rho_M\left(1-\varphi_F\right)}\right]\rho_F}, \tag{2}$$

where ρ is the density of the polymer composite.

Calculated according to (2), the system porosity values increase from 2.3 % with zero filler content up to 4.1 % for aluminum particle content of 5 %.

5. 2. Features of thermal conductivity of polyvinylidene fluoride-filler systems

One of the main thermal characteristics of composite materials is thermal conductivity. For PCM, thermal conductivity depends both on the structure and physical condition of the polymer matrix, as well as on the content, shape, and nature of the filler input. The PCM thermal conductivity level is quite low since the main contribution to the overall thermal conductivity is given by the polymer matrix ($\lambda \approx 0.1$ W/(m·K)). It is known that the PCM thermal conductivity depends not only on the thermal conductivity of the filler [21], its concentration, and its tendency to form its own structures in the composite material but also on the nature of its interaction with the polymer matrix [22].

Fig. 3 shows experimental results of the dependence of thermal conductivity coefficient on the content of filler for composites based on PVDF. Fig. 3 demonstrates that the concentration dependence of the thermal conductivity coefficient is nonlinear. A similar dependence of thermal conductivity was observed by the authors of work [21] for polypropylene-CNTs and polytetrafluoroethylene- CNTs systems, as well as the authors of work [23] for epoxy polymer-Ni and epoxy polymer-Cu systems.

Our analysis of absolute values of the thermal conductivity coefficient has established that the introduction of 5 % Al leads to an increase in the thermal conductivity of the system, based on PVDF, by 10 times.

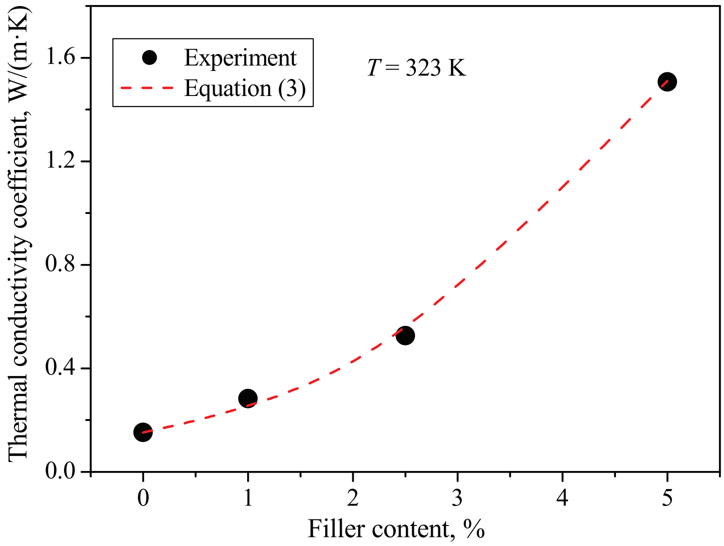

Fig. 3. Dependence of the coefficient of thermal conductivity on the content of the filler for systems based on polyvinylidene fluoride

Unlike "S"-like dependencies obtained and modeled, for example in [24], the thermal conductivity of the PVDF-Al system shows the dependence of another type. For this type of dependencies, an equation was proposed to describe the thermal conductivity of the composite, with the random distribution of the filler [25], where the values of the content of the matrix phase and filler are contained in the degree indicator:

$$\lambda_c = \lambda_r^{(1-\varphi)} \lambda_f^{\varphi}, \qquad (3)$$

where λ_c, λ_r, λ_f – thermal conductivity of composite, polymer matrix and filler, respectively; φ – concentration of filler in the composite.

To calculate thermal conductivity by equality (3) for the thermal conductivity of the matrix, its experimentally obtained value λ_r=0.17 W/(m·K). The thermal conductivity of the filler was determined in the modeling results. For the SYSTEM PVDF-Al λ_f was 25 W/(m·K). Fig. 3 shows that the thermal conductivity values calculated for (3) are in good accordance with experimentally defined values for the PVDF-Al system.

5. 3. The influence of fillers on the values of temperature transitions in polyvinylidene fluoride-based systems

To study the effect of fillers on the values of temperature transitions, the obtained materials were investigated by the method of differential scanning calorimetry. We have derived the dependences of the specific heat capacity on temperature $C_p(T)$. The experimental results are shown in Fig. 4.

All DSC curves of the studied systems demonstrate a maximum in the temperature range of 430–460 K, which is responsible for the melting of the material, as well as a sharp drop in heat capacity in the temperature range of 520–560 K, indicating the beginning of its destruction.

The results of our calculations of melting temperatures (T_m) and destruction (T_d) are shown in Fig. 5. Fig. 5 demonstrates that the melting point and destruction temperature in the case of filling with dispersed aluminum particles depend on the contents of the filler. Both temperatures increase almost monotonously throughout the studied concentration range of the filler. However, this dependence is not quite characteristic of systems such as polymer-filler.

For example, for the polyethylene oxide-nanodiamonds system [26], the melting point showed extreme dependence, which testified to a significant aggregation of filler particles [27]. However, for the PVDF-Al system, this behavior is not observed, which could be indirect evidence of the absence of aggregation in such a system. It should be noted that the heat resistance of the materials under study increases by 20 K with the introduction of 5 % filler, which could have a significant economic effect when using such materials in production.

5. 4. The physical and mechanical characteristics of the polyvinylidene fluoride-filler systems

The peculiarity of polymer composites containing inorganic fillers is the presence of specific viscoelastic

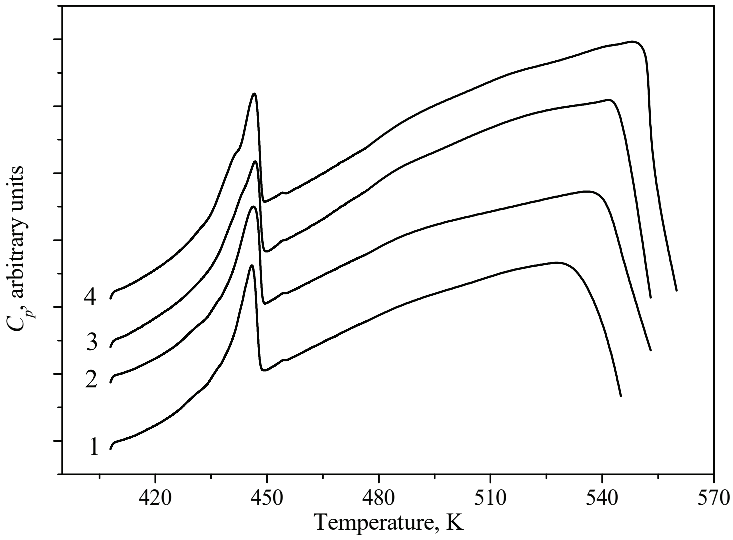

Fig. 4. Dependence of specific heat capacity on temperature for PVDF-based systems. 1 – PVDF; 2 – PVDF +1 % Al; 3 – PVDF +2.5 % Al; 4 – PVDF+5 % Al

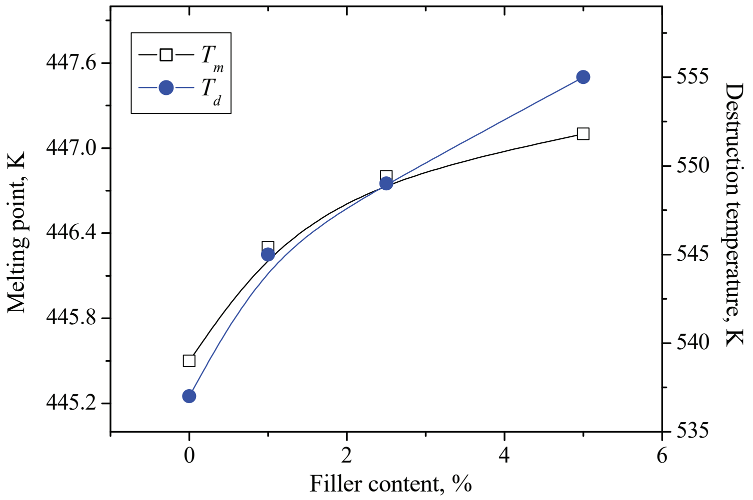

Fig. 5. Dependence of melting point and destruction temperature on the content of the filler for systems based on polyvinylidene fluoride

properties caused by the structure of the material [28]. It is characteristic that they manifest the properties of both elastic bodies and liquids.

The most complete study of the viscoelastic properties of polymer composites, and, accordingly, the effect of the filler on the polymer matrix, could be using acoustic methods. In particular, a change in the velocity and absorption of voltage waves makes it possible to observe relaxation processes, investigate the properties of polymer composites in wide temperature ranges under the influence of heterogeneous factors [28].

To study the effect of fillers on the acoustic characteristics of composites based on PVDF, we studied the features of ultrasound propagation through the samples under study. The results of our research are shown in Fig. 6. They demonstrate that the speed of ultrasound propagation (υ) increases with an increase in the content of the filler, showing almost linear behavior. At the same time, the sound speed increases from 2,120 m/s (for non-filled filled PVDF) to almost 3,400 m/s (for PVDF + 5 % Al), which could be used to produce soundproof materials.

To study the effect of the filler on the mechanical characteristics of the PVDF-Al system, we studied the tensile strength of the obtained materials. Fig. 6 shows the dependence of tensile strength on the content of dispersed aluminum particles. The resulting dependence demonstrates that with an increase in the contents of the filler, the tensile strength of the system increases. At the same time, with a content of 5 % in the material, the tensile strength increases by almost 40 %, which makes this material promising for the creation of structural materials.

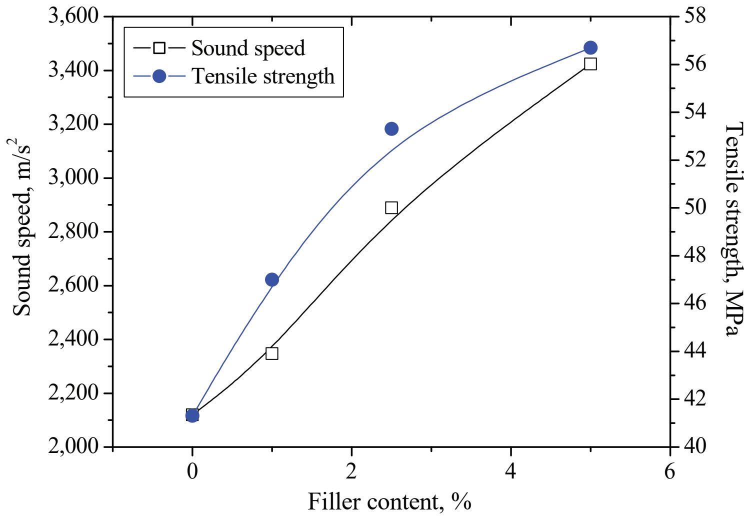

Fig. 6. Dependence of the speed of sound and the module of elasticity on the content of the filler for systems based on polyvinylidene fluoride

6. Discussion of results of studying the influence of the filler on the functional properties of polymer composite materials

The introduction of inorganic filler into the polymer matrix leads to significant changes in its structure, which, in turn, significantly affects the resulting functional properties of the obtained composite material. Such an impact could be observed by analyzing the results shown in Fig. 2. They demonstrate that the experimentally derived density dependences do not coincide with theoretically calculated ones. The deviation in the density, determined experimentally, from the density derived from the law of additivity indicates the direct effect of the filler on the polymer matrix. At the same time, with a decrease in density, compared to the calculated one, we can conclude that the system is loosening. The loosening of the matrix is evidenced by a change in the porosity *P* of the material, which depends on the quantitative content of Al particles. Such an increase in the *P* value indicates that the compression of the polymer matrix is not additive due to the action of the forces of attraction between the macromolecules of PVDF and the active centers of the surface of aluminum particles, as well as the different magnitude of the coefficient of thermal expansion of composite components during their heating [20].

Since the introduction of dispersed aluminum particles leads to the loosening of the matrix, this may indicate the transition of PVDF macromolecules from the crystalline phase to the boundary layer around the filler. This feature of structure formation and the uniform distribution of filler particles with a piston extruder ensured an increase in thermal conductivity of heat resistance, mechanical and acoustic characteristics of the materials obtained in comparison with non-filled PVDF.

The increase in thermal conductivity of PCM (Fig. 3) is associated with the contribution of fillers to the thermal conductivity of the composite, which are characterized by a much higher coefficient of thermal conductivity compared to the polymer matrix [22]. At the same time, the thermal conductivity of pure Al equals ≈235 W/(m·K). At the same time, Fig. 3 also shows that the overall level of thermal conductivity is much lower than the thermal conductivity of pure fillers, even at sufficiently high concentrations of the filler. This can be explained by the fact that the particles of the filler in its pure form are covered with an insulating film. In the case of aluminum particles, it is an oxide film (Al_2O_3), which has a very low thermal conductivity.

According to the authors of work [23], determining the thermal conductivity of the dispersed phase, that is metal powder, is a difficult task, since measuring the thermal conductivity of powders is associated with significant experimental and theoretical difficulties. First, conventional devices are designed to measure solid block materials (or liquids), which makes it impossible to measure powder. Second, the powder is a two-phase system (particles/air), and directly measured values of the thermal conductivity of the powder relate to the thermal conductivity of this two-phase system, and not only to the thermal conductivity of powder particles [23].

The thermal conductivity of metal in the form of powder is significantly less than that for block material [29]. This fact can be explained by the presence of a large thermal resistance at the particle/particle and particle/polymer interface. Thermal resistance impairs the spread of heat flow along the composite and limits the value of its thermal conductivity [30]. The transfer of heat in a filled composite is carried out both through the matrix phase and through the filler phase; at the particle/particle and polymer/particle interface, there is the dissipation of phonons that carry the heat flow [31]. This is evidenced by the results of modeling using equation (3). The value λ_f=25 W/(m·K), derived from modeling, was an order of magnitude less than the thermal conductivity coefficient of block aluminum.

Together with thermal conductivity, the introduction of filler also improves the heat resistance of the materials obtained. The increase in both melting point and destruction temperature is explained by the fact that fillers act as heterogeneous centers of embryo formation. With the increase in the centers of embryo formation of the crystalline phase, both the rate of crystallization and the degree of crystallization of the polymer matrix increase. The result is a more perfect polymer structure with a higher degree of crystallinity than the original [21]. The authors of work [32] also observed a similar increase in the degree of crystallinity of the polymer matrix for the polybutylene terephthalate-carbon nanotubes system.

In addition to thermal properties, modification of the PVDF matrix with the help of dispersed aluminum particles leads to a significant improvement in the mechanical characteristics of the materials obtained. That is why the change in the density of systems based on PVDF, depending on the content of fillers, manifests itself in the viscoelastic and physical-mechanical characteristics of the composite.

The increase in the rate of ultrasound propagation in the obtained materials can be explained by the contribution from the filler, which has greater sound conductivity than the polymer matrix. At the same time, the reinforcing effect of aluminum particles on the polymer matrix is also manifested. Fig. 6 shows that as the filler content increases, the tensile strength of the systems under study increases. This is due to the formation, inside the material, of the reinforcing mesh of filler particles, which, interacting with each other and the matrix, significantly enhance its mechanical characteristics [33].

The prospect of using polymer composites to create structural materials for various purposes is associated with a set of their characteristics, such as the required thermal and mechanical properties, high corrosion resistance [34], relatively small specific weight, etc. [35]. Therefore, it should be noted that the mechanical characteristics and operating temperature range of the examined materials based on PVDF and 5 % Al are much higher than in a non-filled matrix.

This work focused only on determining the effect of dispersed aluminum particles of a certain type on the functional characteristics of polymer composites. The patterns established in the current study cannot be extended to polymer systems that contain a filler of a different type, size, and dispersion.

Further advancement of this study could be to establish more general patterns of influence of fillers of different nature and geometric characteristics on the structure and properties of composites based on thermoplastic polymers. Of significant interest is the construction of theoretical models that would make it possible to predict the resulting properties of polymer composites.

7. Conclusions

1. It has been established that the use of a piston extruder ensures the required level of the distribution of aluminum particles in the PVDF matrix, thereby leading to an increase in thermal conductivity of heat resistance, the mechanical and acoustic characteristics of the materials obtained in comparison with non-filled PVDF.

2. It was found that the introduction of 5 % of fillers leads to an increase in thermal conductivity by almost 10 times. This growth is explained by the contribution from a more thermal conductive filler.

3. It has been shown that the introduction of fillers leads to an increase in the melting point and destruction temperature of PVDF-based systems, which is a consequence of the formation of a reinforcing mesh from filler particles in the polymer material. In this case, the heat resistance increases by 20 K.

4. It has been demonstrated that the physical and mechanical characteristics of the PVDF-based material significantly increase with the introduction of 5 % Al. The sound conductivity increases by 1.5 times, and the mechanical tensile strength – by about 40 %, due to the reinforcing effect of the filler.

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