CNT-Filled Polypropylene/Plasticized Polyvinyl Alcohol Mixtures: Rheology, Morphology, and Properties of Composite Threads

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The influence of the concentration of plasticizer (glycerine) and carbon nanotubes (CNTs) on the micro- and macrorheological properties of melts of polypropylene/plasticized polyvinyl alcohol (PP/PVA) mixtures is studied. During flow of melts of all the studied compositions, a microfibrillar structure is formed within them. As found, changing the ratio of viscoelastic properties of PP and PVA, when adding different amounts of modifying additives, allows us to adjust the mass fraction of types of structures (microfibrils, films, particles), as well as their dimensional characteristics. Reducing the average diameter of microfibrils from 2.7 to 1.2 µm and increasing their share from 84.3 to 96.8 wt.% in the initial and nanofilled mixtures, respectively, are achieved in a composition with similar values of viscosity and elasticity of the components. As shown, the melts of bi- and three-component systems are typical non-Newtonian fluids, and the viscosity of their melts is several times lower than η of the initial components. The values of elasticity, judging by the equilibrium swelling of extrudates, are higher than for the starting polymers, and depend on the content of nanotubes. The ability to longitudinal deformation of the melt jet, which determines its fibre-forming properties, in composite systems is reduced, but remains sufficient for stable processing into threads on available process equipment. As established, the formed microfibrillary structure helps to improve the performance characteristics of composite monothreads due to the effect of self-reinforcement. Tensile strength and dimensional stability of threads obtained from a mixture containing 0.5 wt.% CNTs increase by 1.5 and 1.6 times, respectively.

Вивчено вплив концентрації пластифікатора (гліцерину) та вуглецевих нанотрубок (ВНТ) на мікро- та макрореологічні властивості розтопів сумішей поліпропілен/пластифікований полівініловий спирт (ПП/ПВС). За течії всіх досліджених композицій в них формується мікрофібрилярна структура. Встановлено, що зміна співвідношення в'язкопружніх

властивостей ПП і ПВС через додавання у них різної кількости моди-фікувальних добавок уможливлює реґулювати масову долю різновидів структур (мікрофібрили, плівки, частинки), а також їхні розмірні характеристики. Зменшення середнього діяметра мікрофібрил з 2,7 до 1,2 мкм і збільшення їхньої частки з 84,3 до 96,8 мас.% у вихідній і нанонаповненій сумішах відповідно досягнуто в композиції з близькими величинами в'язкости й еластичности компонентів. Показано, що розтопи бі- та трикомпонентних систем є типовими неньютонівськими рідинами, а в'язкість їхніх розтопів у декілька разів нижча за η вихідних компонентів. Величини еластичности, судячи з показників рівноважного розбухання екструдатів, є вищими, ніж для вихідних полімерів, і залежать від вмісту нанотрубок. Здатність до поздовжньої деформації розтопу струменя, яка визначає його волокноутворювальні властивості, у композиційних систем понижується, але залишається достатньою для стабільної переробки у нитки на наявному технологічному обладнанні. Встановлено, що сформована мікрофібрилярна структура сприяє поліпшенню експлуатаційних характеристик композиційних монониток за рахунок ефекту самоармування. Розривна міцність і формостійкість ниток, одержаних із суміші, що містить 0,5 мас.% ВНТ, зростають в 1,5 і 1,6 рази відповідно.

1. INTRODUCTION

The discovery of carbon nanotubes (CNTs) in 1991 led to significant advances in nanotechnology and marked a new era in the material world, especially in the field of polymer nanocomposites. Single- and multilayer CNT have a complex of unique mechanical, electrical, thermal and chemical properties, as well as a high ability to transfer electrons. The modulus of elasticity of carbon nanotubes is close to the values of this indicator for diamond (1.0 and 1.2 TPa, respectively); their strength is 100 times higher than strength of the best steel samples. They are also characterized by high electrical conductivity ($\sim 10^3$ S/cm), thermal stability (up to 2800°C in vacuum), and thermal conductivity (approximately twice higher than for diamond) [1]. Today, there are various methods of introducing CNTs into polymer matrices, which have made it possible to create new promising materials with multifunctional properties. At this, the modifying effect is manifested with a small amount of filler. Because the ratio of the length of nanotubes to the diameter is from

hundreds to the thousand, high conductivity was achieved even at an additive concentration of 0.0025 wt.% [2]. CNTs/polymer composites are used in a variety of fields: as reinforced and anti-corrosion materials, solar panels, chemical sensors, adsorbents, shielding products from electromagnetic and microwave radiation, *etc.* [1]. Carbon nanotubes with unique graphitic structure, superior optical and biological properties have attracted more and more interests in biomedical applications, including gene/drug delivery, bioimaging, biosensor and tissue engineering [3].

A necessary condition for achieving maximum indicators of properties of nanocomposites is to ensure uniform dispersion and orientation of CNTs in the system, as well as the ability to transfer stresses from the matrix to the filler. Functionalization of CNTs' surface is one of the methods that prevents their aggregation, increases the affinity for polymer melts in the mixture and provides predictable placement in the composition [4, 5]. Modification of carbon nanotubes with titanium stearate allowed reducing their content in the mixture of CNTs/polyolefin by an order compared to the original tubes, and obtaining hydrophobic composites with high hardness and wear resistance [5].

It is known that in mixtures of incompatible polymers highly dispersed fillers are unevenly distributed in the phase volumes of different by chemically nature components. This phenomenon significantly expands the possibilities of regulating of heterogeneous morphology and makes mixed composites even more attractive [6–11]. A review of recent advances in CNTs-filled composites shows that a number of factors can control their functional properties: selective localization of nanotubes (thermodynamic and kinetic effects), changes in morphology due to their compatibilizing action, effects on rheological properties of components and on melting and crystallization processes [6]. Thus, the mechanical and dielectric properties of composites based on an incompatible mixture of polystyrene (PS)/polyvinylidene fluoride were regulated by selective localization of CNTs in one of the phases, using two-stage mixing [7]. A significant increase in the tensile strength of biodegradable composites based on a mixture of poly(ε-caprolactone)/poly(lactic acid) was achieved due to interfacial adhesion and the formation of percolation mesh in the matrix with the simultaneous use of organomodified montmorillonite and multilayer CNTs [8].

Regulation of homogeneity of the morphology of polymer dispersions and selective localization of nanoadditives in one of the phases can also be achieved by introducing an additional component—a compatibilizer [9–11]. The use as a compatibilizer of polystyrene with grafted maleic anhydride in a mixture of PS/polyamide/CNTs has increased the homogeneity in size and geometric shape of PS

drops, regardless of the sequence of injection of components [8]. The materials obtained from these compositions had high electrical, electromagnetic and mechanical properties. In our previous works, it was shown that the preliminary injection of additives CNTs or CNTs/compatibilizer into the melt of a mixture of polypropylene (PP)/copolyamide helps to reduce the average diameter of PP microfibrils and the proportion of unwanted structures (particles, films) [10, 11]. The established regularity is due to the compatibilizing effect of modifiers, which is confirmed by the decrease in the value of the interfacial tension. At the same time, the mechanical characteristics of composite mono- and complex microfibrillary threads obtained from the studied mixtures increase.

Thus, significant progress has been made in the development of polymer/CNTs composites characterized by unique mechanical and physical properties. In order to use fully the properties inherent in carbon nanotubes, it is advisable to conduct further theoretical and experimental studies on the formation of morphology at the macro-, micro- and nanoscale, rheological behaviour of systems and characteristics of finished products.

2. OBJECTS AND METHODS OF RESEARCH

The studies were performed using a mixture of polypropylene/polyvinyl alcohol composition of 30/70 wt.%. The dispersed phase was isotactic polypropylene brand 575P company 'SABIC' with melt flow index of 11 g/10 min (230°C/2.16 kg, ISO 1133-1) and a melting point of 169°C, and the dispersion medium—polyvinyl alcohol brand Moviol 5–88 company 'Kuraray Co. Ltd' with the following characteristics: viscosity of 4% aqueous solution at a temperature of 20°C—5.5–5.8 mPa·s, the degree of hydrolysis—86.7–88.7%, the content of volatile compounds—5.0 wt.%. The flow and destruction temperatures of PVA are very close that makes it impossible processing through the melt. To solve this problem, it was plasticized with glycerine, which was injected in an amount of 10.0 wt.% (PVA$_1$) and 15.0 wt.% (PVA$_2$). As the nanoadditive were selected a three-layer carbon nanotubes manufactured by 'Spetsmash LTD' (Ukraine) with the following characteristics: bulk density—29 g/dm^3, specific surface after acid cleaning—340 m^2/g, outer diameter of nanotubes—10–20 nm. The concentration of CNTs in mixtures was 0.1–1.0 of wt.%. The compositions were prepared on the worm-disc extruder ('DniproPolymerMash', Ukraine). Carbon nanotubes were pre-introduced into the PP melt, and then, the obtained granules were mixed with plasticized PVA. The rheological properties of the melts, *i.e.*, viscosity (η) and flow mode (n), were investigated on the capillary microviscosimeter at a temperature of 190°C in the range of

shear stress τ $(0.10-5.69) \cdot 10^4$ Pa. Construction of flow curves and calculations were made using specially created software [12].

The elasticity of melts was determined by the value of the equilibrium swelling of extrudates (B) [13]. The ability of the melts to longitudinal deformation was evaluated by the value of the maximum degree of drawing (F_{max}), which was calculated as the ratio of the jet reception velocity to the linear velocity of the melt in the spinneret. Composite threads were formed on a laboratory stand through a die with a diameter of 780 µm with a degree of drawing of 1000%. Thermoorientational drawing was performed at a temperature of 150°C with a multiplicity of 4.2–5.0. The tensile strength, the initial modulus, *i.e.*, the force required to deform the thread by 1.0%, and the relative tensile elongation of the composite threads were determined using a rupture machine of KT 7010 AZ brand. The microstructure of extrudates was studied by optical microscopy. Digital micrographs of cross-sections of extrudates and PP residues after PVA extraction were obtained using a digital camera with an adapter. Quantitatively structure formation was assessed by determining under a microscope MBR-15 all types of PP structures and their sizes. Experimental data were processed using software, and the average diameters of microfibres and particles (d) and the mass fraction of each type of structures were calculated [12].

3. RESULTS AND DISCUSSION

Studies of the rheological properties of the starting polymers have shown that the introduction of nanoadditives and plasticizers differently affects the effective viscosity of the melts of PP (η_1) and PVA (η_2) and the nature of their flow (Tables 1, 2).

Increasing the glycerine content by 5.0 wt.% causes a decrease in η_2 of the PVA melt in $\cong 1.5$ times, and with increasing concentration of CNTs from 0.1 to 1.0 wt.%, viscosity of polypropylene increases by 1.1–1.6 times. In this case, the degree of deviation from the Newtonian regime of the flow of melts of plasticized polyvinyl alcohol decreases, and for the composition of PP/CNTs, it increases.

The decrease in the value of the indicator (n) for the PVA sample with a higher concentration of glycerine is natural, because the plasticization occurs by intrabundle mechanism. There is a destruction of the supramolecular structure of the polymer, which brings its flow regime closer to the Newtonian fluid. CNTs additives structure the melt that is accompanied by an increase in the viscosity anomaly.

The viscosity of the melts of PP/PVA mixtures is reduced by 2–3 times compared to η of the starting ingredients (Table 2). Glycerine is a selective plasticizer and remains in the mixture in a thermody-

TABLE 1. The effect of glycerine and CNTs additives on the rheological properties of PVA and PP melts[*].

Name of polymer	Additive, wt.%		Viscosity, Pa·s	Flow mode	Swelling	Maximum deformation, %
	glycerine	CNTs				
PVA_1	10,0	0	490	1,5	1,3	14900
PVA_2	15,0	0	330	1,3	1,4	12700
PP	0	0	290	2,0	1,8	26900
PP	0	0,1	310	2,0	1,7	27100
PP	0	0,5	350	2,1	1,5	27900
PP	0	1,0	450	2,1	1,5	28100

[*]at $\tau = 5.69 \cdot 10^4$ Pa.

TABLE 2. The effect of CNTs additives on the rheological properties of melts of mixtures PP/PVA[*].

Sample name	CNTs content, wt.%	Viscosity, Pa·s	η_1/η_2	Flow regime	Swelling	B_1/B_2	Maximum deformation, %
PP/PVA_1	0	150	0.59	1.4	2.2	1.38	8700
PP/PVA_1	0.5	240	0.71	1.7	2.5	1.15	11700
PP/PVA_2	0	110	0.88	1.3	2.6	1.29	8400
PP/PVA_2	0.1	130	0.94	1.5	2.9	1.21	9300
PP/PVA_2	0.5	190	1.06	1.5	3.3	1.07	11400
PP/PVA_2	1.0	230	1.36	1.6	2.8	1.07	11600

[*]at $\tau = 5.69 \cdot 10^4$ Pa.

namically advantageous phase of PVA.

The sharp drop in the viscosity of the composition may be due to the process of structuring the component of the dispersed phase in the matrix. Isolation of PP in a separate phase with the formation of a continuous surface between the components helps to reduce the viscosity due to the sliding of polymers on the phase separation surface.

At the same time, the flow mechanism changes, namely, there is a transition from the segmental movement of polymer macromolecules in the flow to the stratified flow of individual components [14].

For nanofilled melts of mixtures, there is an increase in their viscosity at all investigated concentrations (Table 2). At this, the values of η of the three-component compositions remain much lower than the additive values. Since the filled polymer melts are suspensions, their properties are determined by the possibility of interaction of the additive particles with each other, as well as with the ingredients of the mixture. The thickening effect of CNTs in the PP/PVA melt is obviously due to their ability to form network structures [1, 6]. PP/CNTs and PP/PVA/CNTs melts are typical non-Newtonian fluids (Fig. 1, Table 2). At this, the effect of deformation speed on the effective viscosity of the compositions is much smaller than on the η of polypropylene. On the dependence curves $\lg\eta = f(\lg\tau)$ for mixtures of all compositions, the 'structural' section is more inclined than in curves for the original and filled PP. The nature of flow of the studied systems, judging by the values of n, is due to the polymer of dispersion medium that is established for many pairs of polymers [14].

The elasticity of PVA melts also depends on the concentration of the plasticizer, judging by the values of equilibrium swelling of the extrudates: with increasing glycerine content, the value of (B_2) increases slightly. At the same time, the elasticity of nanofilled PP melts (B_1) decreases with increasing amount of additive (Table 1, 2).
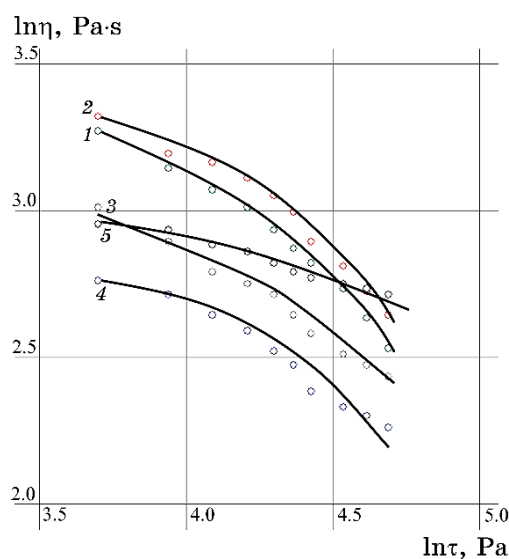

Fig. 1. Dependence of melt viscosity on shear stress: *1*—PP; *2*—PP/CNTs; *3*—PVA$_1$; *4*—PP/PVA$_1$; *5*—PP/PVA$_1$/CNTs.

This is due to changes in the mobility of macromolecules PP and PVA and the possibility of their transition from a spherical ball to elongated chains in the presence of a solid nanofiller and plasticizer, respectively. As can be seen from Table 2, for melts of bi- and three-component mixtures, the increased elasticity is characteristic, compared with the same indicator for the original components. Additional accumulation of large normal stresses occurs due to the deformation of the droplets of the dispersed phase into ellipsoids of rotation and jets (cylinders).

Studies have shown that changes in the viscoelastic characteristics of ingredients in the presence of modifying additives significantly affect the course of microrheological processes during flowing of melts of mixtures PP/PVA. Thus, the degree of dispersion of PP in the PVA matrix depends on the content of the plasticizer. As its concentration increases, the size and geometric shape of the drops change, *i.e.*, their diameters and the proportion of elongated drops decrease (Fig. 2). The injection of carbon nanotubes into the system contributes to the formation of a finer and more homogeneous morphology.

Microscopic studies of PP residues after extraction of the matrix component from the extrudates of all compositions showed that microfibrillar morphology is formed during their flowing. Polypropylene is mainly in the form of microfibrils, as well as films and small particles (Fig. 3). The formation of films is facilitated by the migration of the component with lower viscosity to the capillary walls: the smaller the viscosity ratio (η_1/η_2), the greater the share of films (Tables 2, 3). Micron-size particles apparently appear due to the decay of small-diameter jets, which are the most thermodynamically unstable.

The results of the quantitative assessment of the microstructure of the extrudates are presented in Table 3, which shows that the ratio between its different types significantly depends on the content of plasticizer and nanoadditives in the compositions.

With increasing concentration of glycerine and CNTs, the dimensional characteristics of PP microfibrils decrease, and their mass fraction increases. The modifying effect of the nanoadditive depends on its concentration and reaches the maximum at content of 0.5 wt.%, the average diameter of the fibrils is reduced by more than 2 times. One of the reasons for this is the approximation of values (η_1/η_2) and (B_1/B_2) to 1 (Table 2); as it is known, these factors increase the degree of dispersion and deformation of droplets during flowing of melts of incompatible polymer mixtures [14]. The *in situ* formation of PP microfibrils of smaller diameters may also be the result of the compatibilizing action of nanotubes. Reducing the value of surface tension and increasing the stability of thin fi-
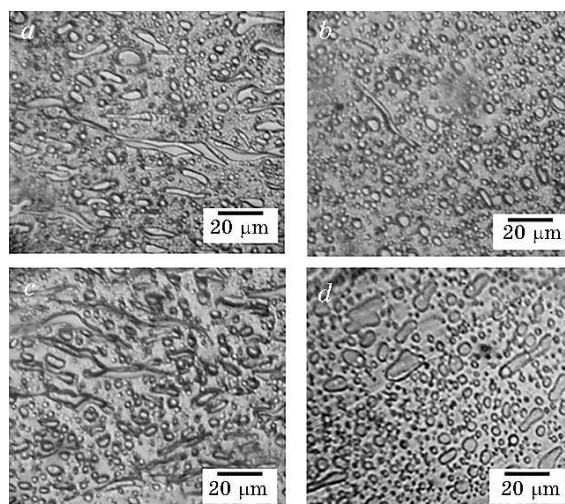
Fig. 2. Microphotographs of cross-sections of extrudates of the initial mixtures PP/PVA and filled with 0.5 wt.% of CNTs: *a*) PP/PVA$_1$; *b*) PP/PVA$_1$/CNTs; *c*) PP/PVA$_2$; *d*) PP/PVA$_2$/CNTs.
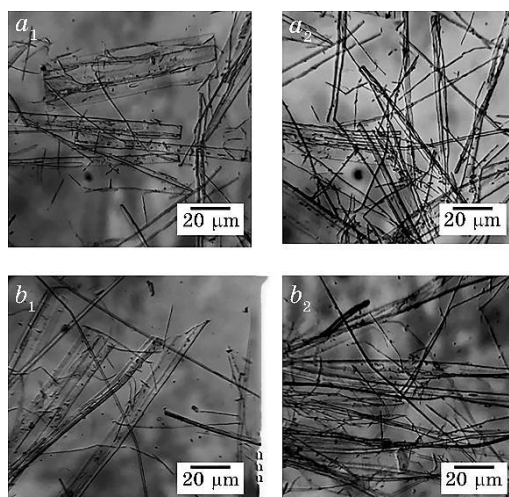

Fig. 3. Microphotographs of polypropylene residues after extraction of the matrix component from the extrudates of mixtures: *a*) the original PP/PVA$_2$ (*a*$_1$—films, *a*$_2$—microfibrils); *b*) PP/PVA$_2$ /CNTs with a content of 0.5 wt.% CNTs (*b*$_1$—films, *b*$_2$—microfibrils).

brils, we found earlier for a mixture of PP/CPA/CNTs [10, 11]. The use of two-stage mixing of components contributed to the migration and selective localization of nanotubes at the interphase area.

TABLE 3. Characteristics of the microstructure of extrudates of the original and nanofilled mixtures.

Name of mixture	Content of CNTs, wt.%	Microfibrils		Particles		Films, wt.%
		d, μm	wt.%	d, μm	wt.%	
PP/PVA$_1$	0	3.4	77.5	3.6	0.7	21.8
PP/PVA$_1$	0.5	1.6	83.0	2.0	0.4	16.6
PP/PVA$_2$	0	2.7	84.3	2.8	0.5	15.2
PP/PVA$_2$	0.1	2.0	90.9	1.8	0.2	8.9
PP/PVA$_2$	0.5	1.2	96.8	1.3	0.1	3.1
PP/PVA$_2$	1.0	1.7	96.0	2.1	0.1	3.9

Regulation of the microstructure of polymer systems by adding CNTs has been described for many incompatible mixtures of polymers [10, 11, 15–19]. Carbon nanotubes can increase the degree of dispersion of the component of dispersed phase or its coarsening, cause the droplets to combine into clusters or in liquid jets, to form continuous structures or domains of irregular shape.

The modifying effect of nanotubes, in addition to the above factors, is associated with suppression of coalescence due to the formation of a solid barrier around small droplets, creating a physical network by them and adsorption interaction of polymer macromolecules with CNTs' surface [15].

Thus, introduction of 1.0 vol.% CNTs into the mixture of PP/polystyrene caused significant changes in the morphology of the mixtures of complementary composition and with a component ratio of 50:50 [19]. At this, in all the studied systems, the nanotubes were mainly localized in the PS phase or at the interphase area. The authors explain the significant morphological changes by the migration of CNTs between domains in combination with their effect on the viscoelastic properties of the components.

It is known that the feature of polymeric liquids is the ability to longitudinal deformation of the jet, flowing from the hole of the die, which led to the very existence as a branch of technology of fibrous materials, in particular chemical fibres. Technological parameter that characterizes the ability of the melt to uniaxial tensile is the value of maximum degree of deformation (F_{max}). The fibre-forming properties of polymer compositions are better the higher the value of F_{max}. Experimental data show that the values of deformation of the melts of the mixtures of PP/PVA in the field of the longitudinal velocity gradient are smaller than those for the original components are. This is due to the heterogeneity of their structure and weak interaction in the transition layer of the segments of macromolecules of polar and nonpolar polymers. The injection of

nanotubes improves the fibre-forming properties of the compositions by increasing the proportion of anisotropic structures (PP microfibrils) with smaller diameters and increasing the viscosity of nanofilled melts. The F_{max} values of bi- and three-component systems remain lower than in the original PP and PVA, and they are stably processed into threads on existing process equipment.

The mechanical characteristics of monothreads formed from the original polymers and modified compositions are given in Table 4.

The analysis of the obtained results shows a significant improvement in the mechanical properties of monothreads containing additives of carbon nanotubes.

The values of tensile strength and initial modulus of threads from the original and modified mixtures are greater than the additive values (Table 4).

The degree of increase in strength and dimensional stability for threads from nanofilled systems is higher than that from polypropylene. The maximum mechanical indicators have threads, in which the prevailing type of structure is microfibrils with the minimum diameters (Tables 3, 4). This is achieved primarily through self-reinforcement, which is characteristic of products with microfibrillar morphology [20]. The improvement of the performance characteristics of monothreads from nanofilled mixtures may also be due to the selective localization of CNTs in the PP phase or at the interphase, because the greater the relationship between the dimensional characteristics of nanoobjects, the faster they move in the system [21]. In addition, according to some authors, this is facilitated by the formation by nanotubes of percolation mesh, 'cross-linked' structure or the effect of nanobridge of CNTs at the interphase area [15].

TABLE 4. The effect of CNTs additives on the mechanical properties of monothreads from PP melts and mixtures of PP/PVA$_2$.

Name of polymer, mixture	Content of CNTs, wt.%	Tensile strength, MPa	Modulus of elasticity, GPa	Elongation, %
PP	0	330	2.6	12.9
PP	0.1	370	2.8	12.6
PP	0.5	450	3.7	12.0
PP	1.0	470	3.8	11.8
PVA$_2$	0	310	2.9	13.7
PP/PVA$_2$	0	340	3.1	14.0
PP/PVA$_2$	0.1	410	3.7	13.9
PP/PVA$_2$	0.5	520	4.9	13.5
PP/PVA$_2$	1.0	530	4.8	13.3

4. CONCLUSIONS

The influence of the concentration of carbon nanotubes on the rheological properties and morphology of melts of polypropylene/plasticized polyvinyl alcohol mixtures with different glycerine content was established. It is shown that the melt viscosity of the initial mixtures decreases by almost 3 times compared to the η of matrix, regardless of the plasticizer concentration. This is due to the formation of liquid jets and PP films in the PVA melt and the formation of a continuous separation surface between them, which facilitates the transition from segmental movement of polymer macromolecules in the flow to the stratified flow of individual components. Increasing η of compositions containing 0.1−1.0 wt.% CNTs is due to the ability of nanotubes to form mesh structures. The melt elasticity of bicomponent and ternary mixtures increases, compared to the same indicator for the initial components, which is due to the additional accumulation of normal stresses by deformed drops of the dispersed phase.

It is shown that the change of viscoelastic characteristics of ingredients in the presence of modifying additives allows regulating the morphology of PP/PVA mixtures. Approximation of the values of the ratio of viscosities and elasticities of the components to 1 in a mixture containing 0.5 wt.% CNTs caused a decrease in the diameters of microfibrils PP from 2.7 to 1.2 μm and the proportion of films and particles from 15.7 to 3.2 wt.%, respectively.

The possibility of self-reinforcement of monothreads from initial and filled with CNTs mixtures of PP/PVA due to the formation of microfibrillar structure in them was established; the value of tensile strength and the initial modulus of the threads are higher than the additive values. Maximum indicators of strength and dimensional stability are achieved provided that the predominant type of structure is microfibrils with minimal diameters. The formation of threads from mixtures, in which the component of the dispersed phase in the flow forms *in situ* microfibrils, is the most attractive, because it simplifies threads production, and the effect of modification becomes much higher.

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