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# INFLUENCE OF CARBON NANOTUBES ON THE CHARACTERISTICS OF POLYMER MELT FLOW DURING EXTRUSION OF TWO-LAYER PIPES

This paper presents the results of numerical simulation of the extrusion process of two-layer polymer pipes in a twodimensional axisymmetric formulation using the ANSYS Polyflow software package. The influence of the introduction of multi-walled carbon nanotubes (MWCNTs) into polypropylene (PP) on the rheological properties of the melt and the flow characteristics in the extrusion head was investigated. The Bird-Carreau model was used to describe the viscosity of the PP/MWCNT composite, taking into account the concentrations of MWCNTs (0, 1, 2, 4, and 8 wt%). The second layer of the pipe was made of polyvinyl chloride (PVC). An analysis of the distributions of pressure, melt flow velocity, and layer thicknesses of the extruded pipe was conducted. It was found that with increasing MWCNT content, the viscosity of the PP/MWCNT composite significantly increases, leading to an increase in pressure in the extrusion head (from 91.3 kPa at 0 wt% to 28.5 MPa at 8 wt%) and affecting the layer thickness distribution: the thickness of the PP/MWCNT layer decreases from 2.49 mm to 1.70 mm, while the thickness of the PVC layer increases from 2.50 mm to 3.45 mm. The optimal concentration of MWCNTs was determined to be 2 wt%, which provides balanced rheological characteristics and stable pipe geometry (total thickness of 5.23 mm). At a concentration of 8 wt%, the PP/MWCNT layer remains intact, but its thickness becomes insufficient for practical use. The obtained data allow optimizing the composition of the polymer nanocomposite and the parameters of the extrusion process for the production of two-layer pipes with specified properties.

*Keywords*: polymer nanocomposites, carbon nanotubes, extrusion, rheology, two-layer pipe, polypropylene, polyvinyl chloride

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#### Introduction

Polymer pipes are widely utilized across various industries, including construction, water supply, gas supply, and agriculture, owing to their corrosion resistance, light weight, and relatively low cost. The production of polymer pipes is predominantly achieved through the extrusion process, during which molten polymer is forced through a forming head that defines the geometry of the final product. The quality and performance characteristics of the pipes are contingent upon the uniformity of the melt distribution at the head's exit, as well as the level of residual stresses that develop during the cooling of the extrudate.

A critical factor influencing the extrusion characteristics and the formation of residual stresses is the design of the extrusion head. Conventional head designs frequently fail to ensure optimal melt distribution, particularly in the extrusion of multi-layer pipes composed of polymers with differing rheological properties. Uneven melt flow results in non-uniform wall thickness, the formation of defects, and a reduction in the pipe's strength properties. Furthermore, the viscosity of the melt, which is dependent on the shear rate, exerts a significant influence on the extrusion process.

In recent years, researchers have increasingly focused on the incorporation of nanofillers, such as multi-walled carbon nanotubes (MWCNTs), to enhance the properties of polymer materials. The addition of MWCNTs to the polymer matrix substantially improves the mechanical, thermal, and electrical properties of the resulting composite. However, the effectiveness of these nanofillers hinges on their degree of dispersion within the polymer matrix. Agglomeration of MWCNTs diminishes their reinforcing effect and may adversely impact the quality of the extruded products.

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Special consideration must be given to the phenomenon of viscous encapsulation when designing extrusion heads for multi-layer pipes. Viscous encapsulation occurs during the co-extrusion of polymers with significantly different viscosities, where the less viscous material tends to flow around the more viscous one. This can lead to uneven layer distribution, alterations in the product's geometry, and even the rupture of one of the layers. This issue becomes particularly pertinent when employing nanofillers like MWCNTs, which can markedly increase the viscosity of the polymer matrix, thereby creating a substantial viscosity disparity between layers and consequently amplifying the viscous encapsulation effect.

## **Analysis of Literary Sources**

The process of extruding polymer pipes, as well as other thermoplastic products, has been widely investigated over many years. In [1], the possibility of refined design of the forming channel geometry in extrusion heads using numerical simulation is demonstrated. The objective of the simulation is to determine the channel shape that ensures a uniform polymer flow distribution at the die exit. The ability to modify the forming channel using an optimization algorithm enables the calculation of the channel geometry for specified dimensions of the extruded product. The ANSYS Polyflow software package for hydrodynamic modeling is employed to develop the extrusion head. Similar studies aimed at optimizing the design of extrusion heads to achieve uniform melt distribution are presented in [2]. In this scientific work, a comparison of two extrusion head configurations for producing pipes from HDPE (high-density polyethylene) is conducted: one with four inlet channels and another with eight. The simulation results revealed that the head with eight inlet channels provides higher pipe quality due to a more uniform melt distribution.

A critical aspect in the design of extrusion heads is the consideration of the rheological properties of the polymer melt. In [3], the rheological behavior of polycarbonate composites with multi-walled carbon nanotubes (MWCNTs) is explored. The authors note that the addition of MWCNTs leads to an increase in melt viscosity. Through oscillatory rheometry, the study demonstrates an increase in melt viscosity in the low shear frequency range (less than 1 rad/s), corresponding to slow material deformations. Comparable findings regarding the influence of MWCNT concentration on the rheological properties of polymer composites are reported in [4] and [5].

In [4], the effect of wall thickness non-uniformity on the distribution of residual stresses in extruded high-density polyethylene pipes is investigated. The authors employ a combination of experimental studies and numerical simulation to analyze the cooling process and the formation of residual stresses. The results indicate that variations in the wall thickness of the polyethylene pipe significantly affect the temperature distribution around the circumference during cooling, which, in turn, leads to the redistribution of residual stresses along the circumferential direction. In [6], the optimization of an extrusion head is presented to achieve a uniform velocity and temperature profile at the exit. The authors utilize the nonlinear Giesekus model to describe the rheological behavior of the polymer melt and perform numerical simulation of the extrusion process using the ANSYS Polyflow software package.

Particular attention in the design of extrusion heads for multi-layer pipes should be given to the issue of viscous encapsulation, which occurs during the co-extrusion of polymers with differing viscosities. This phenomenon manifests as the penetration of a less viscous polymer layer into a more viscous one, deforming it, or conversely, the more viscous polymer flowing around the less viscous one, resulting in uneven layer distribution across the thickness. In the case of extruding two-layer pipes—where the outer layer consists of more viscous polypropylene with carbon nanotubes (PP/CNT) and the inner layer of less viscous polyvinyl chloride (PVC)-encapsulation may lead to the penetration of PVC into the PP/CNT layer or the envelopment of the PVC layer by the PP/CNT layer. In [6], solutions are proposed to enhance technological capabilities and broaden the range of materials suitable for co-extrusion to form multi-layer structures. Viscous encapsulation is minimized by optimizing the design of the feed block, while secondary elastic flows are reduced by improving wall slip through the use of special additives. In [7], researchers demonstrate that the direction of encapsulation is determined not only by the viscosity ratio but also by the jump in the difference between normal stresses acting at the interface of phases with different densities. The authors show that the interface is always convex on the side of the liquid with a greater absolute value of this stress difference. This finding aligns well with the observed deformation of the PP/MWCNT layer at high MWCNT concentrations in this study, where its viscosity significantly exceeds that of PVC. Additionally, in [6], methods to minimize viscous encapsulation and elastic instabilities in multi-layer extrusion are proposed, including the use of a multi-layer feeder, a die with specialized geometry, and the application of external lubricants. Conversely, in [8], the authors highlight the significant influence of normal stress difference on layer deformation during the flow of viscoelastic polymers in non-circular channels.

Thus, the analysis of literary data indicates that designing extrusion heads for the production of multi-layer polymer pipes is a complex task requiring consideration of multiple factors, such as head geometry, rheological properties of the polymers, and extrusion process conditions. The application of modern numerical simulation methods enables the optimization of head design and ensures the required characteristics of the products obtained through extrusion.

### **Problem Statement**

This paper considers the effect of introducing multiwall carbon nanotubes (MWCNTs) into polypropylene (PP) on the flow characteristics of a two-layer polymer melt in an extrusion head. The main objective of the study is to determine the optimal concentration of MWCNTs in PP that ensures uniform distribution of the melt at the exit from the extrusion head and minimizes the parameter of deviation of the layer thicknesses of the two-layer pipe from the specified ones. In addition, the study evaluates the effect of MWCNTs on the melt pressure and the melt flow rate. Using numerical modeling, a detailed analysis of the extrusion process is carried out and the relationships between the MWCNT concentration and the melt flow characteristics are revealed. As a result of numerical experiments, characteristic features of the flow of a two-layer melt in an extrusion head are established at different concentrations of MWCNTs in PP, as well as the concentration of MWCNTs necessary to ensure the most efficient extrusion process of high-quality two-layer polymer pipes with specified geometric parameters.

#### **Finite Element Model and Solution Method**

For modeling the flow of the polymer melt in the extrusion head, the mass and momentum conservation equations were used in an axisymmetric formulation. For an incompressible fluid in cylindrical coordinates  $(r, \theta, z)$ , the continuity equation is expressed as [9]:

$$\frac{1}{r}\frac{\partial}{\partial r}(rv_r) + \frac{\partial v_z}{\partial z} = 0 \tag{1}$$

where  $v_r$  and  $v_z$  – are the radial and axial components of velocity, respectively. The equations of motion (Navier-Stokes) for axisymmetric flow are given by [9]:

For the radial direction (r):

$$\rho\left(\frac{\partial v_r}{\partial t} + v_r\frac{\partial v_r}{\partial r} + v_z\frac{\partial v_r}{\partial z}\right) = -\frac{\partial p}{\partial r} + \mu\left(\frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial}{\partial r}(rv_r)\right) + \frac{\partial^2 v_r}{\partial z^2}\right) + \rho g_r \tag{2}$$

For the axial direction (z):

$$\rho\left(\frac{\partial v_z}{\partial t} + v_r \frac{\partial v_z}{\partial r} + v_z \frac{\partial v_z}{\partial z}\right) = -\frac{\partial p}{\partial z} + \mu\left(\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial v_z}{\partial r}\right) + \frac{\partial^2 v_z}{\partial z^2}\right) + \rho g_z \tag{3}$$

where:  $\rho$  is the density (kg/m<sup>3</sup>), *p* is the pressure (Pa),  $\mu$  is the dynamic viscosity of the fluid (Pa·s),  $g_r$  and  $g_z$  are the projections of the gravitational acceleration vector onto the radial and axial directions (m/s<sup>2</sup>), respectively. In these equations, the terms on the left describe inertial effects (fluid acceleration), while the terms on the right account for pressure, viscosity, and gravitational forces acting on the fluid. Since the polymer melt is a non-Newtonian fluid, the viscosity  $\mu$  in equations (2) and (3) is represented as  $\eta(\dot{\gamma})$ , determined by the Bird-Carreau mode [1].

For the study of the flow characteristics of a two-layer polymer melt in the extrusion head, a three-dimensional geometric model of the internal flow channel was developed using the Design Modeler module of the ANSYS software package. The geometry of the head corresponds to a transverse design with two concentric channels for supplying melts: polypropylene (PP) with multi-walled carbon nanotubes (MWCNTs) for the outer layer and polyvinyl chloride (PVC) for the inner layer.

Figure 1 presents a three-dimensional model of the internal channel of the extrusion head, demonstrating the arrangement of the inlet channels, the forming element, and the outlet opening. For simplicity of calculations and considering the axisymmetric nature of the problem, a part of the three-dimensional model corresponding to the outlet section of the head and the initial section of the extrudate formation was selected.

Based on this, a two-dimensional axisymmetric model was created, which was discretized by finite elements (FE), mainly of the rectangular type.

The thickness of the forming die opening was 5 mm. The resulting FE mesh was imported into the Polyflow module of the ANSYS software package. Figure 2 illustrates the discretization of the cross-section of the studied extrusion head.





Figure 1 - Three-dimensional model of the internal cavity of the two-layer extrusion head.

Figure 2 - Discretization of the cross-section of the studied extrusion head.

For modeling materials, polypropylene (PP) with the addition of multi-walled carbon nanotubes (MWCNTs) was selected for the outer layer, and polyvinyl chloride (PVC) for the inner layer. The rheological behavior of PP with MWCNTs was described using the Bird-Carreau model, with parameters (consistency coefficient  $\eta_0$ , characteristic frequency  $\lambda$  and power-law index n) varying depending on the MWCNT concentration [10]. The rheological behavior of PVC was also described by the Bird-Carreau model, with parameters provided in Table 1. The densities of PP and PVC were set to 1100 kg/m<sup>3</sup> and 1260 kg/m<sup>3</sup>, respectively.

Mathematically, the Bird-Carreau model is expressed as [1]:

$$\eta(\dot{\gamma}) = \eta_{\infty} + (\eta_0 - \eta_{\infty}) [1 + (\lambda \dot{\gamma})^a]^{\frac{1-n}{a}} , \qquad (4)$$

where:  $\eta(\dot{\gamma})$  - is the effective viscosity (Pa·s);  $\eta_0$  - is the zero-shear viscosity (Pa·s);  $\eta_\infty$  - is the infinite-shear viscosity (Pa·s);  $\lambda$  - is the relaxation time (s);  $\dot{\gamma}$  - is the shear rate (s<sup>-1</sup>); *a* - is the parameter that controls the steepness of the transition between the Newtonian and power-law flow regimes; *n* - is the power-law index.

The parameters  $(\eta_0, \lambda, n)$  vary depending on the MWCNT concentration, as shown in Table 1 [10].

РР						DVC
MWCNT content	0 wt%	1 wt%	2 wt%	4 wt%	8 wt%	FVC
ηο (Pa·s)	20778	34214	172783	2.106	2.107	0.31313.107
$\lambda$ (s)	0.207	0.514	0.942	2.645	2.733	1.00.102
n	0.4	0.34	0.29	0.27	0.25	0.291766

#### Table 1- Parameters of the Bird-Carreau Model for PP and PVC [10]

Figure 3 shows graphs of viscosity versus shear rate for PP with varying MWCNT contents (from 0 to 8 wt%) and for PVC. It is evident that the addition of MWCNTs significantly affects the viscosity of PP.

The boundary conditions were defined as follows: at the inlet of each channel, the volumetric flow rate was specified:  $2.76E-06 \text{ m}^3/\text{s}$ , for the inner layer and  $3.06E-06 \text{ m}^3/\text{s}$  for the outer layer.

On the walls of the extrusion channels, the no-slip condition (v = 0) was applied.

The model is axisymmetric about axis C (Fig. 4), so on the axis of symmetry, conditions  $v_r = 0$  and  $\frac{\partial v_z}{\partial r} = 0$  were imposed.

At the end of the free extrudate (point A, Fig. 4), the outflow condition was used, and point B corresponds to the end of the die. For modeling the free surface of the extrudate between points A and B, the free surface condition was applied, which accounts for two main requirements: dynamic condition: The normal stress at the liquid-air interface

is typically taken as zero (traction-free condition) if air viscosity is neglected; kinematic condition: The particles of the fluid on the surface move with it, i.e., the normal component of velocity equals the speed of surface displacement: [11]:

$$v \cdot n = \frac{\partial h}{\partial t'},\tag{5}$$

where h is the function describing the shape of the free surface, and n is the normal to it.





Figure 4 - Cross-sectional view of the modeling domain: (a) region of the initial extrudate section, (b) location of the die end, and (c) axis of symmetry.

Figure 3 - Viscosity of PP with varying MWCNT contents and PVC as a function of shear rate.

For the extrudate region, local mesh refinement (local remeshing) was applied using the adaptive method, which evolutionarily rebuilds the FE mesh with respect to the surface deformation. In this method, mesh nodes are moved along predefined lines to ensure accurate modeling of the extrudate shape

Mathematically, the displacement of an internal mesh node along these lines is described by the parametric equation [11]:

$$x = (1 - \xi)x_1 + \xi x_2 + \xi(1 - \xi)d, \qquad (6)$$

where:  $x_1$  and  $x_2$  are the coordinates of the initial and final points of the line;  $\xi$  is a parameter varying from 0 to 1 along the line; d is a vector determining additional node displacement (typically used for shape correction).

The calculations were performed in a steady-state regime using the finite element method (FEM) with the ANSYS software package.

#### **Results of Numerical Modeling**

Numerical modeling of the flow of a two-layer melt in the extrusion head was conducted to investigate the influence of the concentration of multi-walled carbon nanotubes (MWCNTs) in polypropylene (PP) on the key parameters of the extrusion process. The study encompassed a range of MWCNT concentrations from 0 to 8 wt%, specifically: 0, 1, 2, 4, and 8 wt%.

The obtained data, presented in Figures 5–11, demonstrate how the percentage of nanotubes in the outer layer affects the redistribution of layer thicknesses, pressure, and melt flow rate during extrusion. This information serves

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as a basis for correctly determining the final geometric and physical characteristics of the extruded two-layer polymer pipes.

Figure 5 shows the pressure distribution in the die for pure PP (0 wt.% MWCNTs). The highest pressure values are observed in the melt inlet zone, which is consistent with theoretical predictions. The distribution of the melt flow velocity both in the die and in the extrudate formation zone in the absence of MWCNTs is shown in Figure 6. The peak velocity values are recorded in the narrow part of the internal outlet channel, which emphasizes the dynamics of the process, and the distribution of the PP volume fraction in the die and extrudate for the base case clearly confirms the equality of the layer thicknesses (Figure 11e).



**Figure 5** - Pressure distribution in the extrusion head (0 wt% MWCNTs in PP)

**Figure 6** - Distribution of PP melt flow velocity in the head (0 wt% MWCNTs)

Changes in the layer thicknesses of the extruded pipe depending on the MWCNT concentration are detailed in Figure 7. At 0 wt% MWCNTs, the total thickness extrudate thickness is 4.99 mm, with the polyvinyl chloride (PVC) layer occupying 2.50 mm and the PP/MWCNT layer — 2.49 mm. With the addition of 1 wt% MWCNTs, the total thickness increases by 1.2% to 5.05 mm: the PP/MWCNT thickness increases by 1.2% to 2.52 mm, and PVC — by 0.8% to 2.52 mm. At 2 wt%, the total thickness reaches 5.23 mm, which is 4.8% higher compared to the baseline, with PVC showing a sharp increase of 27.2% to 3.18 mm, while PP/MWCNT layer decreases by 17.7% to 2.05 mm.

The variation of the thickness of the extruded pipe layers with the MWCNT concentration is shown in detail in Figure 7. At 0 wt% MWCNT, the total thickness of the extrudate is 4.99 mm, with the thickness of the polyvinyl chloride (PVC) layer being 2.50 mm and the thickness of the PP/MWCNT layer being 2.49 mm. With the addition of 1 wt% MWCNT, the total thickness increases by 1.2% to 5.05 mm: the thickness of the PP/MWCNT layer increases by 1.2 % to 2.52 mm and that of the PVC layer by 0.8 % to 2.52 mm. At 2 wt%, the total thickness reaches 5.23 mm, which is 4.8 % more than the initial level, with the PVC layer showing a sharp increase in thickness by 27.2% to 3.18 mm, while the thickness of the PP/MWCNT layer decreases by 17.7 % to 2.05 mm.

Next, at 4 % by weight, the total extrudate thickness is 5.16 mm (an increase of 3.4 %), the PVC layer thickness increases by 32.4 % to 3.31 mm, and the PP/MWCNT layer thickness decreases by 25.7 % to 1.85 mm. Finally, at 8% by weight, the total thickness reaches 5.15 mm (an increase of 3.2 %), the PVC layer thickness increases by 38.0 % to 3.45 mm, while the PP/MWCNT layer thickness decreases by 31.7 % to 1.70 mm. These data indicate a clear trend: with the increase of MWCNT concentration, the PVC layer thickness steadily increases, while the PP/MWCNT layer thickness gradually decreases, maintaining the total extrudate thickness within a relative stable range.

The dependence of the maximum pressure at the end of the head on the concentration of MWCNTs is shown in Figure 8. At 0 wt.%, the pressure is 91.3 kPa, and at 1 wt.% it decreases slightly by 4.5 % to 87.2 kPa. However, with a further increase in the percentage of nanotubes, the pressure increases sharply: at 2 wt.% it reaches 3.63 MPa (39.8 times), at 4 wt.% - 6.17 MPa (67.6 times), and at 8 wt.% it reaches 28.5 MPa, which is 312.2 times higher than the initial value. Such exponential growth is explained by the increased viscosity of the PP/MWCNT composite, which complicates its passage through the channels of the extrusion head.

Figure 9 shows the dependence of the maximum flow velocity of the PP/MWCNT and PVC layers at the outlet of the die as a function of the percentage concentration of MWCNTs. At 0 wt.%, the velocity of PP/MWCNTs is 9.27E-03 m/s, and for PVC it is 9.16E-03 m/s. With the addition of 1 wt.%, these values decrease by 2.9 % to 9.00E-03 m/s for PP/MWCNTs and by 2.7 % to 8.91E-03 m/s for PVC. Starting with 2 wt.%, the velocities stabilize: at 2 wt.% and 4 wt.% they reach 1.05E-02 m/s (an increase of 13.3 % for PP/MWCNTs and 14.6 % for PVC compared to

0 wt.%), and at 8 wt.% The velocity of PP/MWCNT decreases slightly by 1.0 % to 1.04E-02 m/s, while for PVC it remains at 1.05E-02 m/s. This indicates a stabilization of the velocity at higher concentrations.

In the final section of the extrudate (Figure 10), the layer velocities at 0% by weight are 8.19E-03 m/s for both materials. At 1% by weight, they decrease by 1.5 % to 8.07E-03 m/s.





Figure 7 - Dependence of extrudate layer thicknesses on MWCNT concentration











At 2 wt%, the velocity of PP/MWCNT drops by 18.6 % to 6.67E-03 m/s, and for PVC — by 18.8 % to 6.65E-03 m/s. At 4 wt% and 8 wt%, the values stabilize within 6.65–6.66E-03 m/s, which is a consequence of changes in viscosity and pressure in the system.

The results of the study of the change in the thickness of the layers in the final section of the extrudate depending on the different percentage concentration of MWCNTs are presented in Figure 11. The figures clearly show a gradual decrease in the thickness of the PP/MWCNT layer with an increase in the concentration of nanotubes and the preservation of the integrity of the structure of the layer package even at a concentration of 8 wt%.



Figure 11 - Thickness distribution of PVC (black) and PP/MWCNT (white) layers at different MWCNT concentrations: (a) 0%, (b) 1%, (c) 2%, (d) 4%, (e) 8% and (f) 0%

The images clearly illustrate the reduction in the thickness of the PP/MWCNT layer with increasing MWCNT concentration, while the layer retains its integrity even at 8 wt%, although its thickness significantly decreases to 1.70 mm.

Figure 12 presents the interface profile between the PP/MWCNT and PVC layers for MWCNT concentrations of 0 %, 1 %, 2 %, 4 %, and 8 %. At concentrations of 0 % and 1 %, the interface profile is nearly straight, indicating a stable and uniform distribution of layers. Starting from 2 %, a distortion of the profile is observed, which becomes more pronounced at 4 % and 8 %. Specifically, at 8 % MWCNTs, the profile shows a significant shift toward the PVC layer, which is associated with the increased viscosity of the PP/MWCNT composite, affecting the nature of the melt flow in the extrusion head.

The change in the tilt angle of the interface line between the PP/MWCNT and PVC layers depending on the MWCNT concentration is shown in Fig. 13. At 0% and 1%, the tilt angle is approximately 0.90 and 0.89 degrees, respectively, indicating an almost horizontal position of the interface line (Figure 13). At 2 %, the angle becomes negative (-1.58 degrees), and at 4 % and 8 % it continues to decrease to -1.89 and -2.25 degrees, respectively. This

trend indicates that with an increase in the MWCNT concentration, the interface tilts toward the PVC layer, which is a result of a change in the rheological properties of the materials and the effect of viscous encapsulation, when the less viscous PVC partially displaces the more viscous PP/MWCNT composite.





**Figure 12 -** Profile of the interface line between PP/MWCNT and PVC layers in the two-layer pipe for different MWCNT concentrations in the local coordinate system

**Figure 13 -** Dependence of the inclination angle of the interface line between layers on the MWCNT concentration in the PP/MWCNT composite

#### **Discussion of Results**

The results presented in this paper show that the addition of MWCNTs to PP significantly affects the production technology of two-layer polymer pipes with nanomodified layers. Taking into account such features of the extrusion process as a change (redistribution) in the thickness of the layers of two-layer pipes, distortion of the profile of the contact interface between the layers, bending of the extrudate and a sharp increase in pressure in the extrusion head depending on the percentage of nanoreinforcement is a mandatory requirement for ensuring the quality of the manufactured products.

Numerical modeling demonstrates that as the concentration of MWCNTs in the PP/MWCNT composite increases, the thickness of this layer decreases, while the thickness of the PVC layer increases, although the total thickness of the two-layer pipe remains relatively constant. For example, at 0 % MWCNT concentration, the thickness of the PP/MWCNT layer is approximately half of the total thickness, but at 8 %, it decreases significantly, and PVC becomes the dominant layer. This redistribution is explained by the phenomenon of viscous encapsulation, which occurs due to the difference in viscosity between PP/MWCNT and PVC. As the MWCNT content increases, the viscosity of the PP/MWCNT composite significantly increases, while the viscosity of PVC remains lower. As a result, the less viscous PVC partially displaces the more viscous PP/MWCNT layer, altering their geometry.

The interface line profile—the isoline that separates the PP/MWCNT and PVC layers in the free extrudate—is an important indicator of the interaction between these layers. At low MWCNT concentrations (0 % and 1 %), the isoline remains almost straight, indicating stable and uniform flow of the melts of both layers. However, at 2 % MWCNT content, the interface profile begins to distort, and at 4 % and especially at 8 %, this distortion becomes significantly pronounced with a noticeable shift towards the PVC layer. The main reason is the increase in viscosity

of the PP/MWCNT composite, which affects the flow characteristics of the melt both in the extrusion head and after exiting it. At high concentrations (e.g., 8 %), the more viscous PP/MWCNT layer is partially enveloped by the less viscous PVC, causing deformation of the interface. Analysis of the isoline inclination angle confirms these observations: at 0 % and 1 % MWCNT, the angle is close to zero, reflecting the horizontal position of the interface; at 2 %, it becomes negative, and at 8 %, it reaches -2.25 degrees, indicating a significant tilt towards the PVC side.

In addition to the interface distortion, bending of the entire two-layer pipe is observed in the free extrudate immediately after exiting the head. At low MWCNT concentrations (0% and 1%), the extrudate bends downward, towards the PP/MWCNT layer; at 2 %, it remains almost straight; and at 4% and 8%, it bends upward, towards the PVC layer. This behavior is related to the difference in flow velocities and viscosities between the layers. For example, at 0 % MWCNT, the flow velocity of PP/MWCNT is 9.27E-03 m/s, and for PVC, it is 9.16E-03 m/s, providing a slight advantage to PP/MWCNT and causing the bend downward. At 2 %, the velocities of both layers equalize to 1.05E-02 m/s, but the thickness of PVC increases to 3.18 mm compared to 2.05 mm for PP/MWCNT, balancing the influence and making the extrudate straight. At 4 % and 8 % MWCNT, despite similar velocities, the significant reduction in PP/MWCNT thickness and the thickening of PVC cause PVC to dominate the flow, leading to an upward bend. This bending correlates with the interface line profile between the layers: at high concentrations, the interface bends towards PVC, enhancing the overall bending of the extrudate.

The increase in MWCNT concentration also significantly affects the pressure in the extrusion head. At 0% MWCNT, the pressure is only 91.3 kPa, while at 8 %, it increases to 28.5 MPa. This is explained by the complication of the flow of the viscous PP/MWCNT composite through the head channels, requiring greater effort to maintain the flow rate. It is also worth noting that the flow velocities at the end of the head increase at medium concentrations (e.g., to 1.05E-02 m/s at 2%), but in the free extrudate, they decrease, which may be related to changes in pressure distribution and rheological effects after exiting the head.

Thus, the analysis of the results allows determining the optimal MWCNT concentration for balanced behavior of the two-layer system. A concentration of 2 wt% MWCNT is the most acceptable, as it achieves a reasonable distribution of layer thicknesses (2.05 mm for PP/MWCNT and 3.18 mm for PVC), the pressure in the head is moderate at 3.63 MPa, and the free extrudate remains straight with minimal interface distortion. Low concentrations (0 % and 1 %) do not provide sufficient influence of the nanotubes on the composite properties, while high concentrations (4 % and 8 %) lead to excessive thinning of the PP/MWCNT layer, significant interface deformation, extrudate bending, and a sharp increase in pressure, which can be critical for certain applications.

Therefore, numerical modeling not only reveals the causes of observed phenomena such as the redistribution of layer thicknesses, distortion of the interface profile, and bending of the extrudate but also allows optimizing the composite composition for specific production conditions, ensuring a balance between the geometric characteristics of the extrudate and the technological parameters of the extrusion process.

#### Conclusion

In this study, the influence of adding multi-walled carbon nanotubes (MWCNTs) to polypropylene (PP) on the flow characteristics of a two-layer polymer melt (PP with MWCNTs and PVC) in an extrusion head for pipe production was investigated using numerical modeling in the ANSYS Polyflow software package. The research covered MWCNT concentrations ranging from 0 to 8 wt%, which enabled the analysis of changes in the rheological properties of the PP/MWCNT composite and their impact on key extrusion process parameters, such as pressure distribution, flow velocity, layer thickness, interface profile, and extrudate curvature.

The following outcomes were achieved:

- 1. Numerical modeling of the two-layer melt flow was performed for various MWCNT concentrations in PP.
- 2. Data were obtained on the distribution of velocity, pressure, layer thicknesses of the extruded pipe, and the volume fractions of the materials.
- 3. The optimal MWCNT concentration (2 wt%) was determined, which ensures balanced flow characteristics, including the alignment of layer velocities, acceptable pressure in the head, and appropriate geometric parameters of the pipe.

The analysis of the modeling results demonstrated that the addition of MWCNTs to PP significantly affects the layer thicknesses, pressure, and flow velocity of the melt. At an MWCNT concentration of 2 wt%, the flow velocities of both layers align at the end of the head, promoting a more uniform melt distribution. Although the thickness of the PP/MWCNT layer decreases to 2.05 mm and the PVC layer increases to 3.18 mm, the pressure in the head is 3.63 MPa, which is an acceptable value. Further increasing the MWCNT concentration leads to a significant increase in the composite's viscosity and pressure in the head (up to 28.5 MPa at 8 wt%), as well as a reduction in the PP/MWCNT

layer thickness to 1.70 mm, which may complicate the extrusion process and negatively impact pipe quality. The results indicate a substantial influence of viscous encapsulation on the formation process of two-layer pipes at high MWCNT concentrations in PP. The limited growth of the PP/MWCNT layer at 8 wt% MWCNT concentration underscores the need for careful alignment of the rheological properties of the components and extrusion process parameters to ensure the production of high-quality two-layer products. Thus, the use of numerical modeling enables the optimization of the composite material composition and extrusion process parameters to produce high-quality pipes with specified characteristics.

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## References

- 1. K. G. Kovalenko, V. I. Sivetskii, and A. L. Sokol'skii (2013), "Design of an extrusion die for plastic profiles," *Chemical and Petroleum Engineering*, vol. 49, no. 9-10, p. 675-678. doi:10.1007/s10556-014-9817-x
- 2. Y. Nie, I. M. Cameron, J. Sienz, Y.-J. Lin, W. Sun (2020), "Coupled thermal-structural modelling and experimental validation of spiral mandrel die", *The International Journal of Advanced Manufacturing Technology*, vol. 111, pp. 3047–3061. doi:10.1007/s00170-020-06183-z
- P. Pötschke, T. D. Fornes, and D. R. Paul (2002), "Rheological behavior of multiwalled carbon nanotube/polycarbonate composites," *Polymer*, vol. 43, no. 11, pp. 3247–3255, doi:10.1016/S0032-3861(02)00151-9
- 4. S. Deveci, B. Eryigit, and S. Nestelberger (2021), "Re-distribution of residual stress in polymer extrusion: An eccentric approach," *Polymer Testing*, vol. 93, p. 106971, doi:10.1016/j.polymertesting.2020.106971
- R. Arrigo, G. Malucelli (2020), "Rheological Behavior of Polymer/Carbon Nanotube Composites: An Overview" Materials, vol. 13, no. 12, pp. 2771, doi:10.3390/ma13122771
- R. Huang, J. Silva, B. A. Huntington, J. Patz, R. Andrade, P. J. Harris, K. Yin, M. Cox, R. T. Bonnecaze, and J. M. Maia (2015). "Co-Extrusion Layer Multiplication of Rheologically Mismatched Polymers: A Novel Processing Route." *International Polymer Processing*, vol. 30, no. 3, 317-330, doi:10.3139/217.2955
- D. Borzacchiello, E. Leriche, B. Blottière, and J. Guillet (2014), "On the mechanism of viscoelastic encapsulation of fluid layers in polymer coextrusion," *Journal of Rheology*, vol. 58, no. 2, pp. 493-512. doi: 10.1122/1.4865817
- P. D. Anderson, J. Dooley, and H. E. H. Meijer (2006), "Viscoelastic Effects in Multilayer Polymer Extrusion," Applied Rheology, vol. 16, no. 4, pp. 198-205. doi: 10.1515/arh-2006-0014
- 9. M. de' Michieli Vitturi (2025), "Navier-Stokes equations in cylindrical coordinates," [Online]. Available: https://demichie.github.io/NS\_cylindrical/ (accessed Mar. 29, 2025).
- F. Thiébaud, J.C. Gelin (2010)."Characterization of rheological behaviors of polypropylene / carbon nanotubes composites and modeling their flow in a twin screw mixer" Composites Science and Technology, vol. 70, no. 4, pp. 647, doi:10.1016/j.compscitech.2009.12.020
- 11. "ANSYS Polyflow Users Guide PDF," [Online]. Available: https://www.scribd.com/document/445926962/ANSYS-Polyflow-Users-Guide-pdf (accessed Mar. 29, 2025).

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# ВПЛИВ ВУГЛЕЦЕВИХ НАНОТРУБОК НА ХАРАКТЕРИСТИКИ ТЕЧІЇ РОЗПЛАВУ ПОЛІМЕРУ ПІД ЧАС ЕКСТРУЗІЇ ДВОШАРОВИХ ТРУБ

У цій роботі представлено результати чисельного моделювання процесу екструзії двошарових полімерних труб у двовимірній осесиметричній постановці з використанням програмного комплексу ANSYS Polyflow. Досліджено вплив введення багатостінних вуглецевих нанотрубок (БВНТ) у поліпропілен (ПП) на реологічні властивості розплаву та характеристики течії в екструзійній голівці. Для опису в'язкості композиту ПП/БВНТ використовувалася модель Берда-Карро з урахуванням концентрацій БВНТ (0, 1, 2, 4 та 8 % мас.). Другий шар труби виготовлено з полівінілхлориду (ПВХ). Проведено аналіз розподілів тиску, швидкості течії розплаву та товщини шарів екструдованої труби. Встановлено, що зі збільшенням вмісту БВНТ в'язкість композиту ПП/БВНТ суттево зростає, що призводить до зростання тиску в екструзійній голівці (від 91,3 кПа при 0 % мас. до 28,5 МПа при 8 % мас.) та впливає на розподіл товщини шарів: товщина шару ПП/БВНТ зменшується з 2,49 мм до 1,70 мм, тоді як товщина шару ПВХ збільшується з 2,50 мм до 3,45 мм. Оптимальна концентрація БВНТ становить 2 % мас., яка забезпечує збалансовані реологічні характеристики та стабільну геометрію труби (загальна товщина 5,23 мм). При концентрації 8 % мас. шар ПП/БВНТ залишається цілісним, але його товщина стає недостатньою для практичного використання. Отримані дані дозволяють оптимізувати склад полімерного нанокомпозиту та параметри процесу екструзії для виробництва двошарових труб із заданими властивостями.

**Ключові сло**ва: полімерні нанокомпозити, вуглецеві нанотрубки, екструзія, реологія, двошарова труба, поліпропілен, полівінілхлорид

## Список використаної літератури

- 1. K. G. Kovalenko, V. I. Sivetskii, and A. L. Sokol'skii, "Design of an extrusion die for plastic profiles," *Chemical and Petroleum Engineering*, vol. 49, no. 9-10, p. 675-678, 2013. doi:10.1007/s10556-014-9817-x
- Y. Nie, I. M. Cameron, J. Sienz, Y.-J. Lin, W. Sun, "Coupled thermal-structural modelling and experimental validation of spiral mandrel die", *The International Journal of Advanced Manufacturing Technology*, vol. 111, pp. 3047–3061, 2020. doi:10.1007/s00170-020-06183-z
- 3. P. Pötschke, T. D. Fornes, and D. R. Paul, "Rheological behavior of multiwalled carbon nanotube/polycarbonate composites," *Polymer*, vol. 43, no. 11, pp. 3247–3255, 2002, doi:10.1016/S0032-3861(02)00151-9
- 4. S. Deveci, B. Eryigit, and S. Nestelberger, "Re-distribution of residual stress in polymer extrusion: An eccentric approach," *Polymer Testing*, vol. 93, p. 106971, 2021, doi:10.1016/j.polymertesting.2020.106971
- R. Arrigo, G. Malucelli, "Rheological Behavior of Polymer/Carbon Nanotube Composites: An Overview" Materials, vol. 13, no. 12, pp. 2771, 2020, doi:10.3390/ma13122771
- R. Huang, J. Silva, B. A. Huntington, J. Patz, R. Andrade, P. J. Harris, K. Yin, M. Cox, R. T. Bonnecaze, and J. M. Maia. "Co-Extrusion Layer Multiplication of Rheologically Mismatched Polymers: A Novel Processing Route." *International Polymer Processing*, vol. 30, no. 3, 317-330, 2015, doi:10.3139/217.2955
- D. Borzacchiello, E. Leriche, B. Blottière, and J. Guillet, "On the mechanism of viscoelastic encapsulation of fluid layers in polymer coextrusion," *Journal of Rheology*, vol. 58, no. 2, pp. 493-512, 2014. doi: 10.1122/1.4865817
- P. D. Anderson, J. Dooley, and H. E. H. Meijer, "Viscoelastic Effects in Multilayer Polymer Extrusion," Applied Rheology, vol. 16, no. 4, pp. 198-205, 2006. doi: 10.1515/arh-2006-0014
- 9. M. de' Michieli Vitturi, "Navier-Stokes equations in cylindrical coordinates," [Online]. Available: https://demichie.github.io/NS\_cylindrical/ (accessed Mar. 29, 2025).
- F. Thiébaud, J.C. Gelin."Characterization of rheological behaviors of polypropylene / carbon nanotubes composites and modeling their flow in a twin screw mixer" Composites Science and Technology, vol. 70, no. 4, pp. 647, 2010, doi:10.1016/j.compscitech.2009.12.020
- 11. "ANSYS Polyflow Users Guide PDF," [Online]. Available: https://www.scribd.com/document/445926962/ANSYS-Polyflow-Users-Guide-pdf (accessed Mar. 29, 2025).