The Rheological Properties of LDPE Nanocomposites per a Capillary Rheometer: An Experimental and Numerical Study

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Abstract. Rheological properties are very significant in determining processing performance. The increase of only a few percent of nanoparticles by weight will result in substantial improvement in rheological properties. This work focuses on studies of shear viscosity, shear stress and pressure at a shear rate range of (3-1500) s⁻¹ and temperature of 170 °C. The flow properties of low-density polyethylene melt and nanocomposite experimentally studied in a single-bore capillary rheometer with a length-to-diameter ratio of (10:1) and numerically using POLYFLOW-ansys version 15.0 software based on the power-law model. The experimental results show that the viscosity of nanocomposites decreased while the shear stress increased with the shear rate increase. The pressure drop decreases with the shear rate increases for all additional ratios. The experimental and numerical studies of polymer nanocomposites in a capillary die, the shear viscosity, shear stress and pressure behaviour numerically reveal good agreement with the corresponding experimental results.

Keywords: Capillary Rheometer, Viscosity, Shear Stress, Pressure, LDPE nanocomposite, ANSYS / POLYFLOW

1. Introduction

Numerical simulation, an extremely efficient technique, can well be predicted by a complicated phenomenon. However, most recent researchers are still aiming to solve simplified numerical examples, including the hypothesis of material parameters and characteristics of the flow domain. It may be appropriate for numerical algorithms to be established. However, from an industrial point of view, it is essential to regulate entire flow patterns by using a more accurate model for practical flow problems [1]. The analysis of (shear viscosity, shear stress and pressure) in polymer melts is of great importance in many polymer processing operations and has been the subject of interest, both in experiments and in modelling. Recent developments in numerical computation of generalised models, integral models and differential models to solve viscoelastic constitutive equations have enabled simulated predictions of both the entry and exit flow of polymer melts. Therefore, the importance of numerical simulation in predicting the stress distribution and the extrudate swell has grown with experimental data [2].

In the last few years, POLYFLOW has been developed into a sophisticated numerical package for steady polymer flow, dependant on finite element numerical techniques [3,4]. There are three POLFLOW fluid flow analysis systems available in Workbench: The Fluid Flow (POLFLOW) system
provides the full simulation capabilities of POLFLOW; the Fluid Flow-Blow Moulding (POLFLOW) system provides only the application-specific capabilities of POLFLOW that are suited to blow-moulding simulations; and the Fluid Flow-Extrusion POLFLOW) system provides only the application-specific capabilities of POLFLOW that are suited to extrusion simulations [4].

Numerical studies of the extrusion process and moulding are conducted in different numerical and software work areas. For example, the ANSYS program is a collection of programs that contain many specialisations. The ANSYS POLYFLOW-based finite element method is designed primarily to simulate applications where flexible viscous and viscous flows play important roles. The flows may be dependent on isothermal or non-isothermal, two- or three-dimensional, stable or time elements. A software instrument equation, governing the conduct of this element that solves them all, creates a comprehensive construction of how the system acts completely. These outcomes are then presented in tabulated or graphic forms. This type of analysis is typically used for the design and optimisation of a system that would otherwise be too complex to analyse by hand. A system that might fit into this class is one that is difficult owing to its geometry, scale or principal equation [5].

POLYFLOW is primarily used in polymer and rubber processing, food rheology, glass furnaces and many other rheological applications to solve flow problems [6]. POLYFLOW’s finite element numerical technique includes originally solving the equations for a Newtonian fluid [10]. Then, a parameter of evolution is implemented which gradually transfers the constitutive Newtonian equation to that of the viscoelastic type (generalised model) selected. Using POLYFLOW software, numerical simulations were carried out. POLYFLOW is a commercial finite (version 15.0) for which Crochet and his team created an element package [3, 4].

2. MATERIALS USED

The products used by Subic Company (Iran) under the trade name (HP7022) are low-density polyethylene pellets widely used in the extrusion process; LDPE properties include [(MFR=1.92g/10min at 190°C and 2.16 kg), density=0.91g / cm³, Mw=79,200g.mol⁻¹]. LDPE melts are known to be very sensitive to temperature and pressure compared to LLDPE and HDPE. Zinc oxide (ZnO) nanoparticles were supplied by (Shijiazhuang Sun power Technology Co., Ltd, China) and had a density of 5.6 g/cm³ and a diameter mean of d=30–40 nm, and silver (Ag) nanoparticles were supplied by (Intelligent Materials, NANOSHEL Pvt., Ltd, India) and had a density of 10.5 g/cm³ with particle size about 20nm.

3. Measurement of Rheological Properties by Capillary Rheometer

A capillary rheometer is a device intended to assess shear viscosity and other characteristics of rheology (= flow) [7]. Capillary rheometers as shown in Figure 1 are piston die systems that are intended to assess the viscosity of polymer melts as a result of temperature and deformation rates. They can test fundamental polymers, compounds and multiple composites with small strengthening particles or fibres, metal injection moulding feedstock and similar materials [8]. The capillary is the small cylinder channel through which the materials flow during the experiment. It is defined by a length and diameter, and the ratio of the two is named L/D. It is machined by the die, and sometimes the L/ D is immediately referred to the die [9]. The temperature, sample, shape and preheating parameters refer to the indication given for the basic rheological test.
4. Main Assumptions

The capillary die flow model can be simulated with the following main assumptions:
1. There is no slip between the molten polymer and the inner surface of the die
2. A 2D model is used for capillary simulation
3. The melted polymer is incompressible
4. A fully developed, steady, isothermal and laminar flow
5. The viscosity is dependent on the shear rate

The power law model was used to check the non-Newtonian viscosity between experimental and numerical according to the following formula:

$$\eta = k (\dot{\gamma})^{n-1}$$  \hspace{1cm} (1)

The finite element numerical technique used by POLYFLOW involves solving the equation’s non-Newtonian fluid. The ANSYS program/ POLYFLOW based on the finite element technique was used to simulate the flow in a capillary rheometer [10, 11]. The geometry of the model was created using the ANSYS program (POLYFLOW, 2D planer). First, the researchers determined (inlet) fluid entry area, (wall) reservoir and die, the free surface as shown in Figure 2.A. Second, the researchers determined the contact area between the end of the die and the free surface through Figure 2.B. Third, the researchers applied the mesh to the finite element method to divide the model into several elements and nodes, as shown in Figure 2.D regarding sizing of mesh (fine) (5569 nodes and 5304 elements).
5. RESULTS AND DISCUSSION

5.1 Experimental Result

5.1.1 Shear Viscosity

The shear viscosity, shear stress, and pressure were measured over the shear rate range through the one die at a temperature of 170°C. The numerical results exhibited good agreement with the experimental results; therefore, comparisons for shear viscosity, shear stress and pressure were performed quantitatively. The difference between the numerical and the experimental behaviour occurred due to the input data, which is produced from experimental results and calculation. These data are applied in the simulation program as an average value for a certain boundary condition. Figure 3 shows the comparison between experimental and numerical shear viscosity of nanocomposites at different concentrations and additives of (ZnO and Ag) in a capillary die for L/D =10 and 170°C. The experimental shear viscosity indicates lower values compared with its numerical test at a high shear rate. It was clear to understand that the viscosity of the LDPE melt was more sensitive to temperature due to the structure of the chain. The increasing temperature was attributed to an increase in intermolecular interactions, which affected the amount of free volume available between molecular chains of the polymer [12]. Therefore, as the viscosity of the LDPE melts decreased, the flow became easier through the capillary die; then, the pressure drop decreased [13].
Figure 3. Experimental and numerical viscosity curve of LDPE melt and nanocomposites in a capillary die at a) 1% ZnO b) 3% ZnO c) 5%ZnO.

The difference between these two cases converged gradually toward a high shear rate due to the stable input experimental data there. Figure 4 presents the effect of the addition of Ag nanoparticles on the general viscosity behaviour.
Figure 4. Experimental and numerical shear viscosity curve of nanocomposites in a capillary die at (a) 1% Ag (b) 3% Ag (c) 5% Ag.

5.1.2 Shear Stress

The shear stresses of nanoparticle systems were studied for a capillary rheometer with a diameter of 1 mm at 170°C as functions of the shear rate. It can be seen that nanocomposites showed pseudoplastic conduct as shear stress improved in a nonlinear way with shear rate. Figure 5 illustrates that shear stress increases for all additional concentrations’ experimental and numerical tests. The numerical behaviour again indicates higher values compared with the experimental results, due to the average input data selected from the real test.
Figure 5. Experimental and numerical shear stress curve of nanocomposites in a capillary die at temperature $170^\circ$C a) 1% ZnO b) 3% ZnO c) 5% ZnO.

Figure 6. Experimental and numerical shear stress curve of nanocomposites in a capillary die at temperature $170^\circ$C (a) 1% Ag (b) 3% Ag (c) 5% Ag.
5.1.3 Pressure Drop

The pressure drop of LDPE nanocomposite increased with shear rate increases. Figures 7a, 7b and 7c show the test and simulation pressure comparisons. Under the current shear rate, the influence of shear rate on pressure is evident, and the results of the experiments are lower than those obtained through simulated results.

There are two reasons why simulated outcomes may differ: on the one hand, there is the difference between the mathematical model and the practical processing conditions, such as the die-length-to-diameter ratio, the non-slip hypothesis whose influence is ignored. On the other hand, there is the inherent defect of the constitutive equation, which cannot correctly describe polymer rheological characteristics. Quantitatively predicting field variables directly through experimental measurement is still difficult. However, this can be achieved both qualitatively and quantitatively through numerical simulation technology, thereby revealing the mechanism of material forming.
Figure 7. Experimental and numerical pressure curve of nanocomposites in a capillary die at temperature 170°C, (a) 1% ZnO (b) 3% ZnO (c) 5% ZnO d) 1% Ag e) 3% Ag f) 5% Ag.

6. NUMERICAL RESULT

6.1 Shear Viscosity

Figures 8 and 9 visualise the 2D steady viscosity, shear rate contours of LDPE melt and nanocomposites in a capillary rheometer at 170°C, L/D=10, and shear rate (3-800) s⁻¹. At the centre of the capillary die, the viscosity reaches a maximum value, gradually reducing towards the die wall; at the same time, shear rate indicates opposite behaviour at the top of the capillary die: the pressure reaches a maximum value. These contours were compatible with the quantitative viscosity curve.
Figure 8. Simulation of LDPE nanocomposite in a capillary rheometer of L/D = 10, shear rate A) 3 s$^{-1}$
B) 800 s$^{-1}$ a) 1% ZnO b) 5% ZnO.
**Figure 9.** Simulation of LDPE nanocomposite in a capillary rheometer of L/D =10, shear rate A) 3 s$^{-1}$ B) 800 s$^{-1}$ a) 1% Ag b) 5% Ag.

### 6.2 Pressure and Shear Rate Relation

According to Figures 10 and 11, which represent the numerical result of pressure with shear rate, it is very clear that the behaviour demonstrated with pressure value increasing with the increase of the shear rate (3-800 s$^{-1}$) is good agreement with the experimental results of pressure and shear rate. Figure 10 reveals that the mean of numerical simulation and the power law model are very active in predicting the changes in the pressure with the increase of shear rate. This creates a very accurate means by which to predict the value of the increasing value in the pressure, and it is very active in predicting the effect of the different parameters which are affected by the pressure, such as a nano-effect. The results of the model agree with researchers such as Mustafa [14] as well as with experimental results.
Figure 10. Simulation of LDPE nanocomposite in a capillary rheometer of L/D=10 and 170°C at shear rate A) 3 s⁻¹ B) 800 s⁻¹ a) 1% ZnO b) 3% ZnO c) 5% ZnO.
7. CONCLUSIONS

As the shear viscosity, shear stress and pressure were tested experimentally and numerically, this study concluded that, as the shear rate increases, the shear viscosity decreases, while the pressure and shear stress increase. The numerical simulation of the 2D non-Newtonian flow based on the power law model showed good agreement regarding the shear viscosity, both quantitatively and qualitatively. The experimental and numerical study of polymer nanocomposites as non-Newtonian melt indicates a good agreement between each other.

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