Elongational Behavior of Short Glass Fiber Reinforced Thermoplastics

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Abstract. *Composite thermoplastic materials are used increasingly in several industries. In particular, glass fiber reinforcement is used to improve the mechanical properties of thermoplastics. However, few data of material properties of these fluids are avaiable in the literature. In this work, a study of shear and elongational properties of a commercial short glass fiber reinforced polypropylene is presented. The shear and elongational viscosities were obtained using the pressure drop measured at a capillary rheometer, with axisymmetric converging dies. Two different die geometries were used: semihyperbolically convergent dies and conical convergent dies. In the last case, the elongational viscosity was obtained using the Cogswell and Binding analysis. Numerical simulations were also performed, to investigate the flow field through the extrusion die process, and to evaluate the pressure drop and elongational viscosity. The conservation equations of mass and momentum were solved via the finite element method, using the commercial program POLYFLOW (Ansys). The Oldroyd B constitutive equations were used to model the viscoelastic mechanical behavior of Polypropylene, but the comparison between numerical results and experimental data obtained from the capillary rheometer did not show good agreement.*

Keywords: *capillary rheometer, elongational viscosity, viscoelastic flows, composite materials*

1. INTRODUCTION

Composite thermoplastic materials, such as polypropylene reinforced with short glass fibers, are used increasingly in the automotive and other manufacturing industries to substitute conventional materials. One of its advantages is that they can be shaped in conventional plastics processing equipment such as extruders and injection molding machines. Moreover, glass fiber reinforcement improves the mechanical properties of thermoplastics. The mechanical properties of the final parts depend strongly on the process itself, on the rheological properties of the filled polymer and on the fibers orientation during processing.

The characterization of polymer melts and solutions in elongational flow fields is still a major challenge. Since extension is present in most types of flow processes, such as extrusion and injection molding, shear measurements are not able to describe polymer behavior in this kind of flows. Therefore, it is important to obtain other material properties, as the elongational viscosity, so that one can build and test models and theories, as well as confirms numerical simulations of several flow processes. Despite the importance of elongational measurements, elongational rheometry has lagged behind shear rheometry, due to the a variety of difficulties encountered in the former, such as the difficulty to generate a steady and controlled elongational flow field, the difficulty to compensate for any shear effects which may simultaneously occur, and difficulties in sample production (Collier *et al.*, 1998). Different techniques were developed to obtain the elongational viscosity. One of the most used method is the one that uses die entrance flows. In this method, the elongational viscosity is obtained by measurements of pressure drop and flow rate through the die. Cogswell (1972) and Binding (1988) were the ones that first developed this technique, and since then several authors discussed and presented some improvements to these technique. However, these methods are not able to predict the Trouton ratio at low elongational rates due to the assumed flow kinematics. Zatloukal *et al.* (2002) showed that the results of the viscosity strongly depend on the shape of the steady elongational viscosity and on the L/D ratio of the orifice die. They have proposed an "effective entry length correction", which improves the capability of the entrance techniques to exactly predict elongational viscosity at low elongational rates. This correction helps to evaluate elongational rheological data from capillary measurements. Feigl *et al.* (2003) presented a generalization of the method of measuring the elongational viscosity using semihyperbolic dies. The authors confirmed that purely elongational flow is produced within the die if wall slip is assumed. Numerical results were obtained and compared to the ones predicted by the constitutive equation. A very good agreement was obtained for a wide range of elongational rates. Collier *et al.* (2005) used the semihyperbolically converging die, and different Hencky strains, to analyze the behavior of elongational viscosity for polymers. They found that an increase of the Hencky strain leads to an increase of the elongational viscosity, and this effect decreases slightly as the elongational strain rate increases.

The shear and elongational properties of short glass fiber reinforced polypropylene using commercial rheometers were studied by Mobuchon *et al.* (2005). The steady-state and complex viscosities of the composites were found to be fairly close to that of matrix, but Cox-Merz rule was not verified for the composites at high rates. The elasticity of the composites was found to be equal to that of the polypropylene matrix. The apparent elongational viscosity was obtained from the pressure drop in the planar converging die using the analysis proposed by Cogswell (1972) and Binding (1988). The elongational viscosity of the polypropylene was found to be much larger than the shear viscosity at low strain rates with a Trouton ratio of about 40 that decreased rapidly with increasing strain rate down. The elongational viscosity of the composites was also found to be close to that of the matrix, with 35 an 5% larger for the 30 and 10 wt% reinforced polypropylene.

The main goal of this work is to report shear and elongational properties of a short glass fiber reinforced polypropylene at large strain rates. The elongational and shear viscosity data were obtained using a capillary rheometer. A numerical investigation is also performed, using the Oldroyd B constitutive equation to model the neat polypropylene. The numerical results were compared to the experimental ones to evaluate the capability of this model to predict the matrix behavior.

2. EXPERIMENTAL ANALYSIS

The viscosity data was obtained using an ACER 2000 capillary rheometer. Neat polypropylene and a 10 and a 30 wt% short glass fiber reinforced polypropylene were supplied in pellet form by Basell Polyolefins. The 10 wt% composite corresponds to the semi-concentrated regime whereas the 30 wt% is in the concentrated regime. All tests were performed at 200^0C. A thermal stabilizer, the Irganox 225, was mixed to the sample to avoid thermal degradation. The stabilizer concentrations were equal to 1% of the weight sample.

The shear viscosity data was obtained using axisymmetric converging conical dies, with entrance angle equal to 60^0, and different aspect ratios (L/D, where L is the capillary length and D is the capillary diameter). The elongational viscosity data was obtained using both conical and semihyperbolically dies, the last ones for two different Hencky Strains, equal to 4 and 7. The Hencky strain is defined by the following equation:

$$\epsilon_h = \ln(R_i^2/R_o^2) \tag{1}$$

where R_i is the intlet diameter and R_o is the outlet diameter. The elongational viscosity is obtained by (Feigl *et al.*, 2003):

$$\eta_E = -\frac{\Delta P}{\dot{\epsilon}\epsilon_h} \tag{2}$$

where ΔP is the pressure drop through the die, and $\dot{\epsilon}$ is the elongational rate (=$\partial v_x/\partial x$).

The elongational viscosity was also obtained from the pressure drop measured using the axyssimetric conical converging die. In this case, the Cogswell (Cogswell, 1972) and Binding (Binding, 1988) analysis were employed. The Cogswell analysis is based on the assumption that the flow is locally fully developed in the converging region. The total pressure drop through the contraction, ΔP, is divided into a shear and a elongational terms. Using the elongational terms, the elongational viscosity is given by:

$$\eta_e = \frac{9}{32}\frac{(n+1)^2}{\eta_w}\left(\frac{\Delta P}{\dot{\gamma_a}}\right)^2 \tag{3}$$

where n is the consistency index of the fluid shear viscosity, modeled by the Power Law equation, η_w is the shear viscosity at the wall, ΔP is the total pressure drop, and $\dot{\gamma_a}$ is the apparent shear rate. Binding (1988) employed a more rigorous theory, using the energy balance, and expressed the the elongational viscosity as a power-law function of the elongational rate:

$$\eta_e = l\dot{\epsilon}^{(t-1)} \tag{4}$$

where l and t were obtained from the pressure drop versus flow rate data.

2.1 Experimental Results

Figure 1 shows shear viscosity as a function of shear rate for the neat polypropylene, 10% and 30% short glass fiber reinforced polypropylene. The results show a shear thinning viscosity behavior. Concerning glass fiber concentration, the shear viscosity slightly increases with the addition of short glass fibers, problably due to the fact that at this range of shear rate, the fibers are oriented to flow direction, and so almost no effect on shear viscosity is observed. Figure 2 shows a comparison of the shear viscosity data of the neat and a 30% fiber reinforced polypropylene, with the results obtained by Mobuchon *et al.*, 2005. The data are in good agreement with the shear viscosity data of Mobuchon *et al.* , 2005. For convenience, the experimental data for the neat polypropylene is multiplied by 0.1.

The experimental elongational viscosity obtained using the semihyperbolic die is shown in figures 3 and 4, for the die's Hencky strain equal to 4 and 7, respectively. It can be observed that the elongational viscosity also decreases with the strain rate, and that the glass fiber has no effect on the elongational viscosity at these ranges of strain rates, also probably due to fibers orientation. Its worth noting that the viscosity data are different for the two geometries, because steady state is not achieved within the dies. This can be observed with the aid of figure 5, which shows another way to represent these data, plotting the viscosity $versus$ residence time, which is defined as $t = \epsilon_h/\dot{\epsilon}$. These curves give the viscosity of the

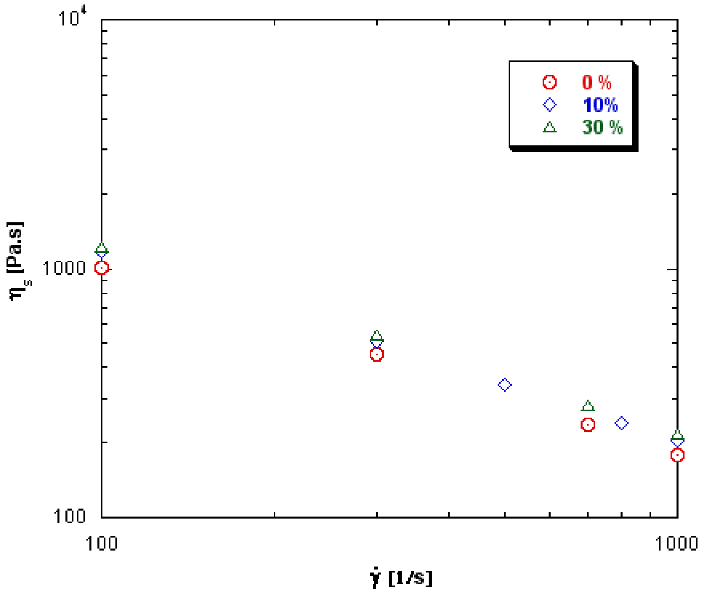

Figure 1. Shear viscosity *versus* strain rate for the neat, 10% and 30% short glass fiber reinforced polypropylene.

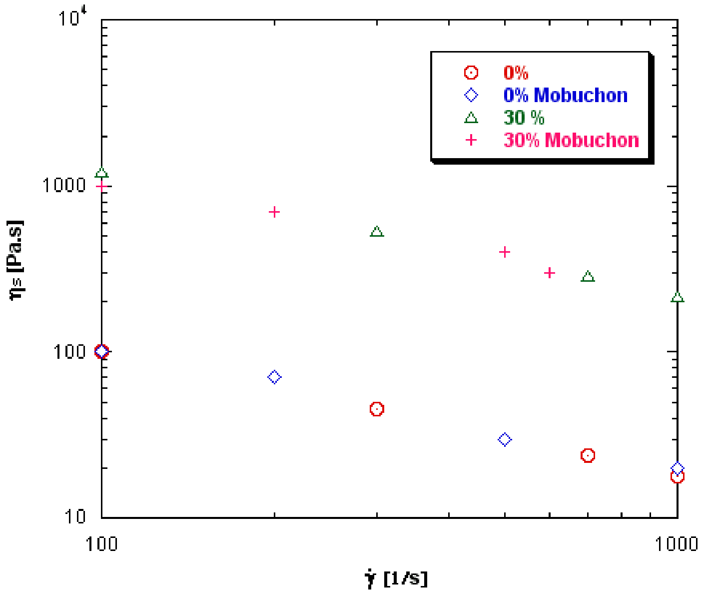

Figure 2. Comparison between neat and 30% short glass fiber reinforced polypropylene with Mobuchon *et al.*, 2005
. The data for the neat polypropylene is multiplied by 0.1 for convenience.

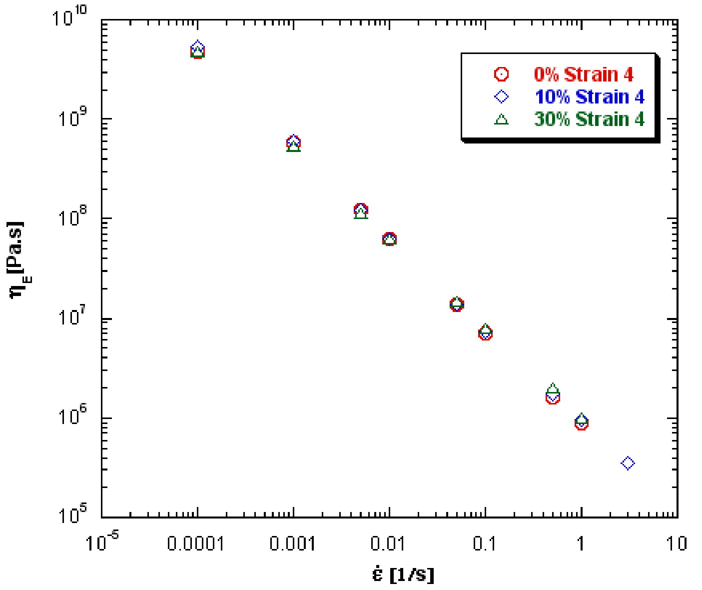

Figure 3. Elongational viscosity data obtained with the semihyperbolic die, Hencky strain 4.

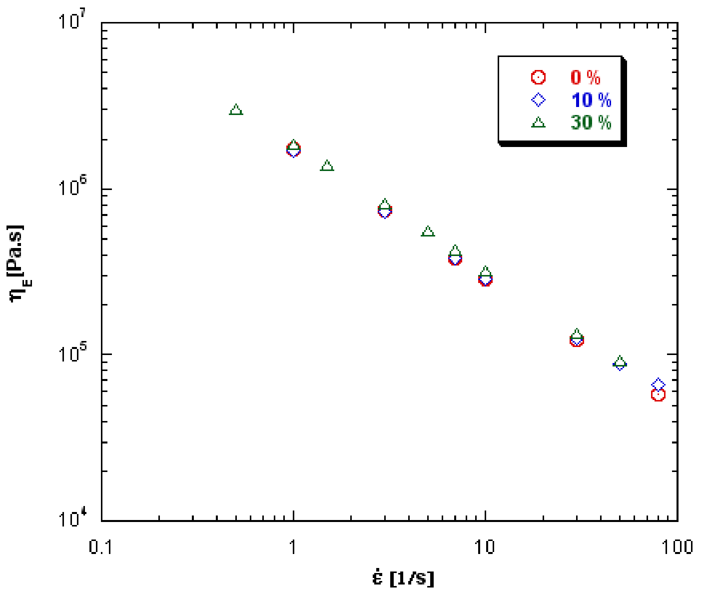

Figure 4. Elongational viscosity data obtained with the semihyperbolic die, Hencky strain 7.

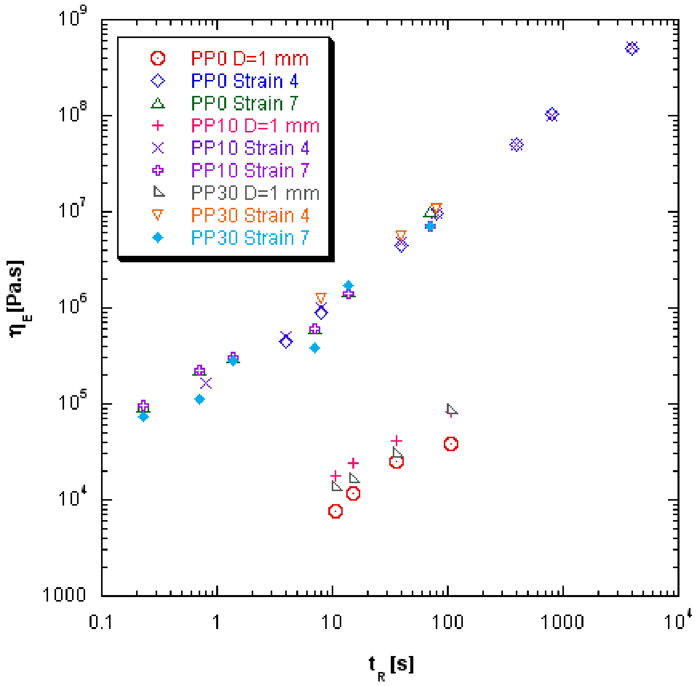

Figure 5. Elongational viscosity data versus time obtained with the semihyperbolic die.

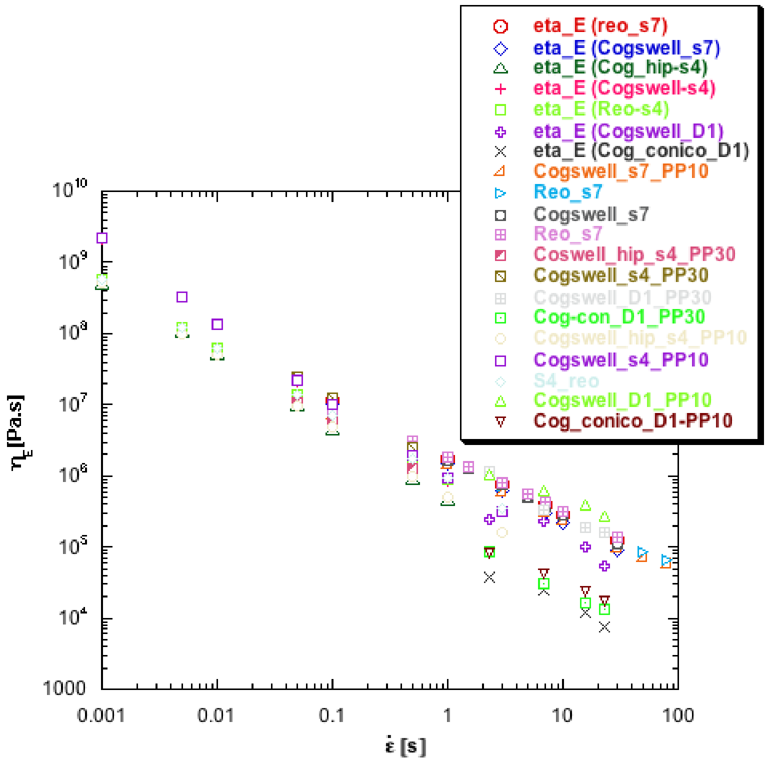

Figure 6. Elongational viscosity comparison.

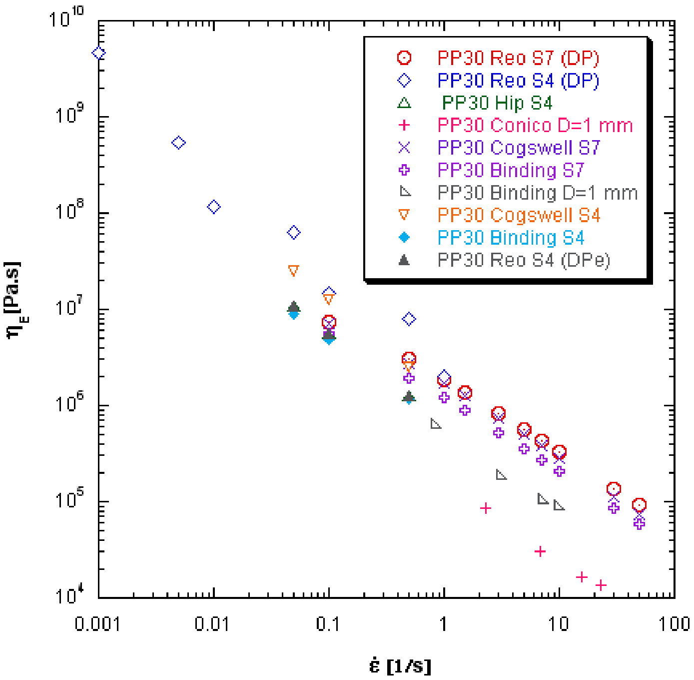

Figure 7. Elongational viscosity comparison.

polypropylene after applying a certain strain to it, and each data point in a certain curve corresponds to a different strain rate. It is noted the linear curve behavior, and that the viscosity values are slightly lower for strain equal to 4. The steady state would be obtained if the polymer largest relaxation time is much lower than the die residence time (which is equal to $t_R = \epsilon_h/\dot{\epsilon}$, Fiegl *et al.* 2003). For this polymer, and using the Oldroyd B model, the relaxation time was estimated as 0.125 s (Bird et al., 1987). The residence time varies with the geometry. It can be seen, with the aid of figure 5, that the residence time varies from 0.8 to 4×10^3 s, for $\epsilon_h = 4$, and from 0.7 to 70 s, for $\epsilon_h = 7$.

Finally, figure 6 shows a comparison of the elongational viscosity results, using different geometries. The results obtained with the conical die were determined via the Cogswell and Binding analysis. It can be observed that all the results are qualitatively similar, but there are some quantitative difference, probably due to the fact that the fluid does not achieve the steady state within the dies, and to the assumptions taken at the Cogswell and Binding analysis. The results obtained with the Binding analysis predict the higher elongational viscosities for the three polypropylene (neat, 10% and 30%), which is in accordance to Zatloukal *et al.* (2002). Figure 7 shows a comparisson of the different assesements to elongational viscosity using the 30% fiber reinforced polypropylene. The results shows that the Cogswell analyses using the semi-hyperbolic geometry strain 4 is equal than the result obtained from the rheometer. However, this behavior is not noticed to strain 7. The possible reason is that the strain 7 geometry seams to a abrupt contraction. An agreement have been noticed from the results of all geometries used and the Cogswell analyses. The results from Binding analyses was higher than the Cogswell analyses in order that the Cogswell assesement the shear contribuition was not used.

3. NUMERICAL ANALYSIS

The numerical analysis was performed to evaluate the flow behavior and to analyze the adequacy of the Oldroyd B model to predict the Polypropylene mechanical behavior.

The commercial software POLYFLOW (Ansys, Inc.) was used to solve the governing equations. POLYFLOW is a finite-element computational fluid dynamics (CFD) program designed primarily for simulating applications where viscous and viscoelastic flows play an important role.

Two different geometries were considered, a convergent axisymmetric conical die, with entrance angle equal to 60^0, and a semihyperbolic axisymmetric die, with the Hencky strain equal to 7. The computational domains for the two cases are shown in figure 9. For the conical geometry entrance die, the radius are $R_0 = 10$ mm, the axial length $L = 20$ mm and the outer radius $R_e = 0.5$ mm. In the second geometry, Hencky strain 7, the entrance radius are $R_0 = 10$ mm, the axial length $L = 25$ mm and the outer radius are $R_e = 0.61$ mm.

3.1 Governing Equations

The numerical solution was obtained solving the conservation equations of mass and momentum, together with an appropriate constitutive equation. The conservation equation of mass is given by:

$$\nabla \cdot \mathbf{v} = 0, \tag{5}$$

The conservation equation of momentum is given by:

$$\rho \left(\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} \right) = -\nabla p + \nabla \cdot \boldsymbol{\tau} + \eta_s \nabla^2 \mathbf{v}, \tag{6}$$

where $\mathbf{v}$ is the velocity vector, ρ is the density, η_s is the solvent (Newtonian) viscosity, p is the pressure, and $\boldsymbol{\tau}$ is the polymeric contribution to the extra-stress tensor.

The mechanical behavior of polypropylene was modeled by the Oldroyd B constitutive equation, given by:

$$\boldsymbol{\tau} + \lambda_1 \overset{\nabla}{\boldsymbol{\tau}} = 2\eta_p \mathbf{D}, \tag{7}$$

where λ is the relaxation time of the fluid and η_p the polymeric viscosity. The upper-convected derivative of $\boldsymbol{\tau}$ is written is terms of

$$\overset{\nabla}{\boldsymbol{\tau}} = \frac{\partial \boldsymbol{\tau}}{\partial t} + \mathbf{v} \cdot \boldsymbol{\tau} - \boldsymbol{\tau} \cdot \nabla \mathbf{v} - \nabla \mathbf{v}^T \cdot \boldsymbol{\tau}, \tag{8}$$

and the rate of deformation tensor **D** is

$$\mathbf{D} = \frac{1}{2}(\nabla \mathbf{v} + \nabla \mathbf{v}^T). \tag{9}$$

The total viscosity, η is written as

$$\eta = \eta_s + \eta_p, \tag{10}$$

where η_s is the solvent viscosity and η_p is the polymeric viscosity.

The relaxation time $\lambda_1 = 0.125$ s was determined from the transient material functions $G^{'}$ and $G^{''}$, obtained experimentally for the neat polypropylene, in Mobuchon *et al.* (2005). The total viscosity used was the one obtained experimentally, which is shown in 1. The ratio between the polymeric and total viscosities was chosen equal to 0.11. The elongational viscosity predicted by the Oldroyd B model in steady and elongational flow is given by (Bird *et al.*, 1987):

$$\eta_E = 3\eta_0 \frac{1 - \lambda_2 \dot{\epsilon} - 2\lambda_1 \lambda_2 \dot{\epsilon}^2}{1 - \lambda_1 \dot{\epsilon} - 2\lambda_1^2 \dot{\epsilon}^2} \tag{11}$$

where $\lambda_2 = \lambda_1 * \eta_p/\eta$. The predicted elongational viscosity is shown in figure 8. It can be observed that it shows a similar qualitative behavior to the experimental results, but the values are underpredicted.

The boundary conditions imposed on the numerical simulations were: flow rate at the inlet, symmetry along the center line, fully developed flow at the outlet, and no-slip conditions at walls.

Mesh tests were performed for the conical geometry. Three different meshes were tested: the first one with 685 triangular elements, and intermediate mesh with 2760 triangular elements and a refined mesh with 6180 triangular elements. The intermediate mesh was chosen, since the differences on the pressure drop through the contraction between this mesh and the refined one, were below 2% . For the semihyperbolic die, a mesh with 3300 triangular elements was used.

3.2 Numerical Results

The velocity, stress and pressure fields for the axisymmetric conical converging die are shown in figures 10-11. Graphs on the left side correspond to $\dot{\gamma} = 1$ s$^-1$, and on the right side to $\dot{\gamma} = 70$ s$^-1$. It can be observed the increasing velocity as the fluid enters the capillary, as expected. The shear stress field shows an increase in the shear stress when the fluid goes through the capillary. It can be also noted that the maximum stress occurs at the wall, as expected. It can be observed, with the aid of figure 11, that the pressure drop through the conical die is of the same order of the pressure drop through the capillary tube. However, for the semihyperbolically die, figure 12, the pressure drop is concentrated near the end of the die. The elongational viscosity was estimated using the results obtained for the semihyperbollically die and eq.2. For $\dot{\epsilon} = 100$ s^{-1}, and using the no slip boundary condition, the numerical solution gives $\eta_E = 5 \times 10^6$ Pa.s, and for $\dot{\epsilon} = 100$ s^{-1} and using slip boundary condition, η_E is almost zero. These values are lower than the ones obtained experimentally, due to the estimated rheological parameters and to the constitutive equation used, which is probably not adequate to predict the polypropylene behavior. The results obtained from the slip conditions have a lower pressure drop, and it improves the elongational viscosiy results.

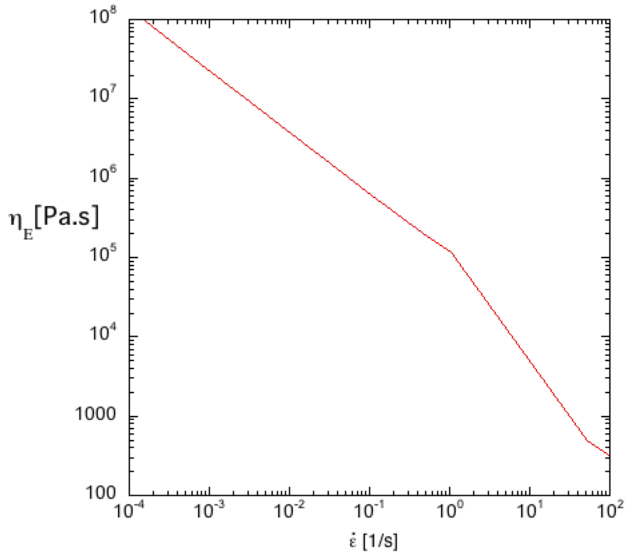

Figure 8. Elongational viscosity predicted by the Oldroyd B model

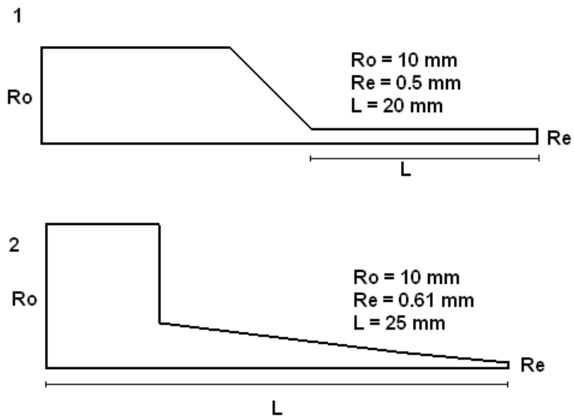

Figure 9. Computational domain for (1) conical and (2) semihyperbollic geometries.

4. CONCLUSIONS

This work presented a study of the shear and elongational viscosities of thermoplastic materials reinforced with short glass fibers. The reinforcement is used to improve the mechanical properties of thermoplastics. The shear and elongational viscosities were obtained experimentally using the pressure drop measured at a capillary rheometer, with axisymmetric converging dies. Two different die geometries were used to obtain the elongational data: conical convergent dies, together with the Cogswell and Binding analysis, and semihyperbolically convergent dies. Numerical simulations were also performed, to investigate the flow field behavior at the extrusion die process, and to evaluate the pressure drop and elongational viscosity. The conservation equations of mass and momentum were solved via the finite element method, using the commercial program POLYFLOW (Ansys), and the Oldroyd B constitutive equations to model the viscoelastic mechanical behavior of Polypropylene.

The experimental results showed that both shear and elongational viscosities decrease with the strain rate, and that the fibers do not affect the viscosity values, due to fiber orientation. The elongational viscosity values were obtained with different methods, and all of them show similar qualitative behavior. However, the results are different because steady state is not reached, and also due to simplified assumptions used in the Cogswell and Binding analysis. The values of the elongational viscosities obtained numerically present the same qualitative trend, but differences were observed, probably due to the Oldroyd B constitutive equation used, which seems to not predict well the polypropylene behavior.

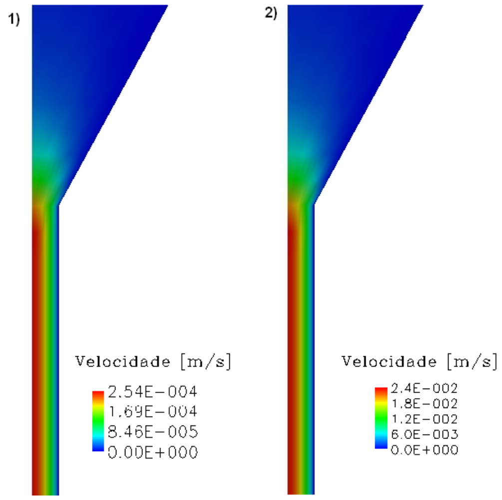

Figure 10. Velocity field in the conical die for (1) $\dot{\gamma} = 1\ \mathrm{s}^{-}1$ and (2) $\dot{\gamma} = 70\ \mathrm{s}^{-}1$.

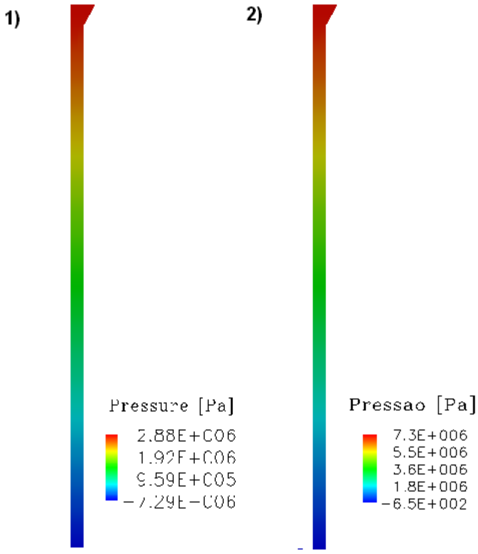

Figure 11. Pressure field in the conical die for (1) $\dot{\gamma} = 1\ \mathrm{s}^{-}1$ and (2) $\dot{\gamma} = 100\ \mathrm{s}^{-}1$.

5. ACKNOWLEDGEMENTS

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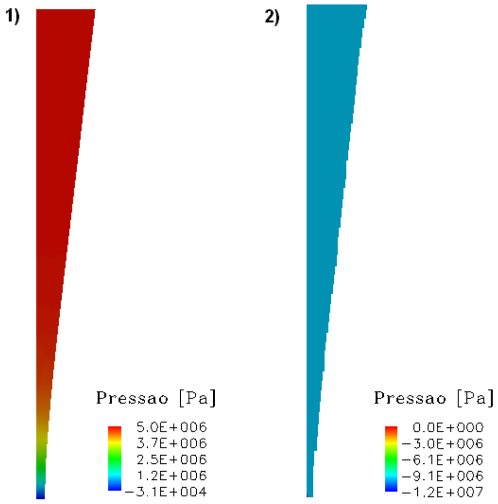

Figure 12. Pressure field in the semihyperbolic die for (1) no slip boundary condition and (2) slip boundary condition.

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