Estimation of injection parameters of a bio-based composite material from the rheological characterization

J I Fajardo, C Paltán, and L M López

1 Universidad Politécnica Salesiana, Cuenca, Ecuador
2 Universitat de Girona, Girona, Spain

E-mail: jfajardo@ups.edu.ec

Abstract. This research work presents an analysis of the injection molding process of a natural fiber-reinforced composite, from the characterization of its rheological properties by the technique of capillary rheometry. Two of the main parameters of injection molding were estimated: the injection pressure and the clamping force. The rheological coefficients obtained were used as inputs in analytical and finite element models, for the prediction of the injection parameters. The results were compared, and it was observed that there is good agreement between them. The percentage errors between the two estimation methods were less than 5% for injection pressure and less than 10% for the clamping force. Also, the behavior of the material using different natural reinforcing contents (compositions of 20%, 30% and 40% fiber with a coupling agent at 4% and 8%) and the effect of its rheological properties on both the injection pressure and clamping force were evaluated. The study is of particular interest given that from the rheological characterization, the experimental values were adjusted to the Cross-WLF viscosity model and the adjustment coefficients, which contain both the reinforcing and coupling agent effects, were fed into the two methods of estimation of the injection parameters. The methodology used and the results obtained allow the prediction of the main parameters of injection molding of new bio-based composites, facilitating their use on an industrial scale.

1. Introduction

The considerable growth in consumption by the world’s population has generated the need to satisfy a great demand for the development of the population. In polymeric materials, this increase in production has been evident, reaching millions of tons of products in recent years [1]. This large consumption harms the environment. Ecological, economic and productive factors have been the basis for establishing the use of biopolymers or bio-based materials as alternatives to reduce this negative impact, making available new materials reinforced with natural fibers to generate responsible consumption and a sustainable future [2–4].

The use of bio-based polymeric materials reinforced with short natural fibers processed by injection molding has gained scientific and industrial interest due to a combination of good mechanical properties and lower environmental impact [5,6]. There has been a remarkable increase in the production of bio-based components worldwide. In Ecuador, we have a wealth of plant species with fiber extraction potential, given the country’s biological richness. Many of these species have not been exploited, so there is wide availability of a variety of plant fibers that can be used as reinforcements of composite materials [7,8]. Advantages of using natural fibers as reinforcement include their biodegradable properties, increased modulus of elasticity (rigidity) and acoustic absorption. They also present environmental advantages in biofuel production.
The injection molding of polymer matrix composites reinforced with natural short fibers is an optimal alternative for the production of a wide variety of products such as sporting goods, decorative items, medical materials and more specific pieces for the automotive field. This manufacturing technique is characterized by its capability for mass production with high dimensional accuracy [9,10].

In the injection molding process, a wide variety of computer assisted engineering (CAE) software is used, allowing the main variables of the process to be estimated. This allows the prediction of the processing conditions and the final product properties [11]. The software lets the user enter the reinforcing material characteristics [11,12]. From a manufacture point of view, it is desirable to predict the processing parameters of these new materials to increase competitiveness by reducing production costs and product development time [11].

The injection process parameters depend on the desired characteristics of the final product. In the filling stage, defects such as welding lines, air bubbles, and incomplete filling can be produced. For these reasons, the study of the rheological behavior of the material is necessary. Parameters such as temperature, shear stress, viscosity, pressure, clamping force among others, should be taken into account when using this manufacturing process. Additionally, the mold and injection machine characteristics must be considered [13].

The present article is focused on an analysis of the injection process of a composite material reinforced with natural fibers, from the characterization of its rheological properties. Two of the main parameters of injection molding were estimated: the injection pressure and the clamping force. Analytical and finite element models (FEM) were run to predict the injection parameters of the injected tensile test specimens.

2. Materials and methods
Polypropylene reinforced with bamboo fibers (PP + BF) at different fiber and coupling agent contents provide by the GiMaT research group from the Universidad Politécnica Salesiana were employed. Polypropylene PPH 7060, from Braskem, with a melt flow index (MFI) of 12 g/10 min was used as a polymeric matrix. As a coupling agent, polypropylene-graft-maleic anhydride (MAPP). The Bamboo fiber employed belongs to mesh number 60; they were previously dried at 110 °C for 2 hours [14].

2.1. Capillary rheometry
The rheological test was carried out under the ASTM D3835-16 standard [15]. The coefficients of the different rheological models were determined. A Malvern Rosand RH2200 double-barrel capillary rheometer was used, with all the analysis using a capillary kit with a length/diameter/angle ratio equal to 20 x 1 x90°, where the angle corresponds to the inclination of the fluid inlet. Bagley correction was applied with a capillary kit of 0.25 x 1 x 180°. In all cases, pressure transducers of 10000 psi and 1500 psi were used. Initially, the behavior was analyzed at 190 °C and 230 °C, but the degradation of the fiber at 230 °C did not allow valid results to be obtained. The range of deformation speed was from 10 s⁻¹ to 2000 s⁻¹.

2.2. Simulation of the injection process
The analysis geometry corresponds to a dogbone specimen for tensile testing obtained by injection. The geometry was exported from 3D modeling software. A 2D Domain type of study was used to solve the interactions in the simulation because the geometry does not present pronounced changes in its thickness, thus saving computational resources. The meshing of the geometry was configured with a distance of 3.5 mm between each node. The mesh elements in their totality are triangular, with a total of 2054 elements used. The properties of the composite material were assigned by selecting the Cross-WLF viscosity model [16]. The software was configured to analyze thermal conditioning, filling, compaction and warping. The injection temperature was set as 190 °C for the melting temperature of the material and 45 °C for the surface temperature of the mold. The injection cycle was set to 45 sec. The cooling system was configured using Ø 10 mm water channels. Tool Steel 420SS was used for the mold material. Finally, the BOY 35E injection machine was configured.
2.3. Analytical calculation

For the analytical calculation, V. K. Savgorodny’s Model was used [17]. The structure and power of the injection mechanism are calculated from the mold filling conditions. It is known that the pressure distribution in the mold during the holding pressure stage determines the clamping force necessary to close the mold. The calculation of average pressure in the rectangular mold will be as in Equation (1), where, \(X\), the maximum dimensions of the mold, (m); \(H\), the thickness of the sample to be molded, (m); \(m\) and \(k\), the constants of the flow curve (3):

\[
\xi_{med} = \frac{4.3 \times 10^7 m^k H^k}{X^2} \left[ 1 - \exp \left( 1.342 \times 10^{-9} \frac{X^2}{m^k H^k} \right) \right] + 1.52 \times 10^5 \frac{m^k H^k}{X^2} \left[ 1 - \exp \left( -2.76 \times 10^{-8} \frac{X^2}{m^k H^k} \right) \right] \tag{1}
\]

The \(f\) coefficient is a dimensionless coefficient that depends on the rheological behavior of the compound and is calculated as shown in Equation (2).

\[
f = \frac{10^{5-2k} m^k}{N \cdot S^k} \tag{2}
\]

The clamping force of the mold is given by Equation (3), where: \(\xi_{med}\), is the average calculated pressure of the mold and \(S\), the surface of the sample.

\[
P = \xi_{med} S \tag{3}
\]

Equation (1) was used to calculate the average pressure and Equation (3) for the clamping force from the constants obtained from the curves that describe the rheological behavior of the \(PP + BF\) composite, these rheological constants adjust the experimental data with the rheological models of the power-law and Cross WLF. These constants are used to calculate the injection parameters of the natural composite material in such a way that the real behavior of the new material is considered. The specimen thickness was 0.004008 m, the flow distance was 0.2101 m and a sample surface area was 0.00645 m².

3. Analysis of results

The rheological study was carried out to determine the coefficients of the different rheological models. It was observed that an increased fiber content in the composite material generated viscosity increases at low shear rates. This effect would lead to processing difficulties through extrusion and injection techniques due to a decreasing of the melting flow index. Leão, et al. [18] reported reduction in the MFI from 12 g/10 min to 2 g/10 min for polyethylene + wood flour composites. This behavior could represent a limitation for injection molding in parts with small channels or with reduced flow areas. On the contrary, the increase in shear rate caused the composite material to present a thinning when approaching values of \(\dot{\gamma} = 1100\) s⁻¹, at this point the viscosity is matrix dominated for all compositions as can be seen in “Error! No se encuentra el origen de la referencia.” Capillary rheometry results of neat PP and composites at 20%, 30% and 40% of BF at 190 °C, and high shear rates showed that the effects of fiber content are not significant. This is particularly important because for injection molding processing the most common shear rate values are in the range 1000 s⁻¹ to 10000 s⁻¹. These findings provide the possibility of designing feed systems for injection molds capable of processing these composite materials. The loss of viscosity that occurs when increasing the shear rates is justified by the change in the molecular conformation of the polymer. The molecules modify their relative position, as the speed increases, the molecules will already be modified to offer the least resistance to movement due to the geometrical parameters of the fibers. Similar effects were reported by Rueda, et al. [19] for glass spheres suspended in a thermoplastic matrix at different concentrations. They suggest that the shape of the reinforcements can be considered as non-agglomerating fillers which prevent the formation of
hydrodynamic clusters. Wan, et al. [20] found a similar thinning for HDPE filled with 40% of maple at different test temperatures.

Figure 1. Capillary rheometry results of neat PP and composites at 20%, 30% and 40% of BF at 190 °C.

Figure 2 shows the relationship between the analytical results of the injection pressure in MPa and the values obtained in the simulation. The behavior of the two curves is similar, with the values obtained having minimal approximation errors in the range of 0.16% to 4.87%. The minimal differences are due to the tri-dimensional analysis carried out by the FEM software; however, this is a good approximation to set injection parameters. It was observed that the fiber content increases the injection pressure with similar values obtained in the analytical analysis and the simulation. This is explained by the increase in the viscosity of the polymer when adding fibers, which demands greater injection pressure. Wan, et al. [20] reported that the addition of rice husk fibers into a polyethylene matrix increased the viscosity of the molten composite, therefore the injection pressure was higher than neat polyethylene. Equation (1) used a pressure variation between the input of the mold and the pressure at distance X, this equation uses a sweep with the maximum dimensions of the fluid path. This considers the injection pressure according to the dimension of the sample, providing a good approximation of the results to the simulation. The composite with the MAPP coupling agent presents a decrease in the injection pressure as the MAPP content increases. By decreasing the viscosity of the composite material with the MAPP coupling agent, the material becomes more fluid and fills the cavities of the mold with less pressure due to the decrease in the flow resistance of the material.

Figure 2. Injection pressure at different fiber and couple agent contents. Simulation vs analytical results.
Figure 3 shows the relationship of the analytical results of the clamping force against the value obtained in the simulation. It was observed that the fiber content increases the value of both the analytical and simulation clamping force. The behavior of the two curves is similar in trend but with slight variations in terms of the magnitude. The values obtained in the simulation have minimal approximation errors in the range of 4.32% to 10.02% with respect to the analytical calculations. Equation (3) was used, which considers the injection pressure with the surface of the injected part. This increase of the clamping force is generated by the increase of the injection pressure and the viscosity of the material when fiber content increases; if the injection pressure rises, the force to resist the opening of the mold increases. The composite with the MAPP coupling agent presents a decrease in the clamping force as its content increases. By decreasing the viscosity of the composite material with the MAPP coupling agent, the material becomes more fluid and fills the mold cavities with less pressure, generating a smaller need for the clamping force.

Figure 3. Clamping force, at different fiber and coupling agent contents. Simulation vs analytical results.

4. Conclusion
The present work evaluated the rheological behavior of a bio-based composite material PP+BF at different compositions and compared the injection pressure and clamping force parameters analytically and through FEM. It was observed that the fiber content has a thickening effect on the polymer matrix, due to the formation of nucleation zones that increase the percentage of crystallinity of the polymer. The coupling agent produces a lubricating effect, improving the processing conditions of the composite by bringing the values of shear viscosity to suitable ranges, allowing for processing by extrusion and injection without needing to increase the sections of the feeding channels or the injection pressure requirements. In all the formulated composites, it was observed that at shear rates close to 2000 s⁻¹, the matrix governs the flow conditions. These parameters can be used in finite element software specialized in injection to characterize the flow properties of this kind of material and obtain more consistent simulations with the real phenomena that occur when processing them. These results allow the configuration of optimized injection cycles for the serial production of components from bio-based materials.

The higher the fiber content of the composite, the higher the injection pressure. When a coupling agent is used, the composite material shows a decrease in injection pressure as the content increases. Fiber content also increases the clamping force. However, as seen already with the injection pressure, composite material with the coupling agent exhibits a decrease in the clamping force required as the content of the latter increases. The simulated results show good agreement with analytical analysis, it is useful for prediction of the main parameters of injection molding of new bio-based composites, facilitating their use on an industrial scale.
References

[1] Faruk O, Bledzki A K, Fink H-P and Sain M 2012 Biocomposites reinforced with natural fibers: 2000–2010 Progress in Polymer Science 37 1552

[2] Chung D H, Kwon T H and others 2002 Fiber orientation in the processing of polymer composites Korea-Australia Rheology Journal 14 175

[3] Ospina M, Mejía de Gutiérrez R, Delvasto S, Monzó J, Borrachero M V and Payá J 2009 Modificación de la morfología de la ceniza de cascarilla de arroz por molienda de alta energía y su efecto en las propiedades reológicas de pastas de cemento portland adicionadas Revista Latinoamericana de Metalurgia y Materiales 1 577

[4] Luna P, Lizarazo-Marriaga J and Mariño A 2016 Guadua angustifolia bamboo fibers as reinforcement of polymeric matrices: An exploratory study Construction and Building Materials 116 93

[5] Hatzikiriakos S G and Dealy J M 1992 Wall slip of molten high density polyethylenes. II. Capillary rheometer studies Journal of Rheology 36 703

[6] Hamad K, Kaseem M and Deri F 2011 Melt rheology of poly (lactic acid)/low density polyethylene polymer blends Advances in Chemical Engineering and Science 01 208

[7] Trujillo E, Moesen M, Osorio L, Van Vuure A W, Ivens J and Verpoest I 2014 Bamboo fibres for reinforcement in composite materials: Strength Weibull analysis Composites Part A: Applied Science and Manufacturing 61 115

[8] Gañán P, Cruz J, Garbizu S, Arbelaitz A and Mondragon I 2004 Stem and bunch banana fibers from cultivation wastes: Effect of treatments on physico-chemical behavior Journal of Applied Polymer Science 94 1489

[9] Yamaguchi H and Fujii T 2004 Bamboo Fiber Reinforced Plastics Natural fibers, plastics and composites (Boston: Springer) pp 305

[10] Pashkova V, Wloch E, Mikolajczyk A, Laniecki M, Sulikowski B and Derewiński M 2008 Composite SBA-15/MFI Type Materials: Preparation, Characterization and Catalytic Performance Catalysis Letters 128 64

[11] Santos J D, Fajardo J I, Cuji A R, García J A, Garzón L E and López L M 2015 Experimental evaluation and simulation of volumetric shrinkage and warpage on polymeric composite reinforced with short natural fibers Front. Mech. Eng. 10 287

[12] Xie J and Jin Y-C 2016 Parameter determination for the Cross rheology equation and its application to modeling non-Newtonian flows using the WC-MPS method Engineering Applications of Computational Fluid Mechanics 10 111

[13] Abbes B, Ayad R, Prudhomme J C and Onteniente J P 1998 Numerical simulation of thermoplastic wheat starch injection molding process Polymer engineering and science 38 2029

[14] Mera-Moya V, Fajardo J I, de Paula Junior I C, Bustamante L, Cruz L J and Barros T 2018 Semi-automatic Determination of Geometrical Properties of Short Natural Fibers in Biocomposites by Digital Image Processing Proceedings of the International Conference on Information Technology & Systems (ICITS 2018) vol 721, ed Rocha A, Guarda T (Cham: Springer) pp 387

[15] American Society for Testing and Materials (ASTM) 2016 Standard test method for determination of properties of polymeric materials by means of a capillary rheometer, ASTM D3835-16 (USA: American Society for Testing and Materials)

[16] Cross M M 1965 Rheology of non-Newtonian fluids: A new flow equation for pseudoplastic systems Journal of Colloid Science 20 417

[17] Savgorodny V K and Uralde L 1978 Transformación de plásticos vol 1 (Barcelona: Gustavo Gili)

[18] Leão A L, Fernandes Teixeira R M and Ferrão P C 2008 Production of Reinforced Composites with Natural Fibers for Industrial Applications – Extrusion and Injection WPC Molecular Crystals and Liquid Crystals 484 157/[523]-166/[532]

[19] Rueda M M, Auscher M-C, Fulchiron R, Périd T, Martin G, Sonntag P and Cassagnau P 2017 Rheology and applications of highly filled polymers: A review of current understanding Progress in Polymer Science 66 22

[20] Wan Abdul Rahman W A, Sin L T and Rahmat A R 2008 Injection moulding simulation analysis of natural fiber composite window frame Journal of Materials Processing Technology 197 22