

Rheological behavior of recycled polypropylene for reuse in the automotive sector

S. FERRÁNDIZ, J. LÓPEZ, R. NAVARRO, F. PARRES

Polytechnic University of Valencia, Materials Technologic Institute, Alcoi – Spain

This work shows the study of rheological behavior of a material that recycled materials have been incorporated. The transformation process to be applied is the injection molding. The mix of materials used was of the same nature and have similar viscosities. This mixture provides virgin polypropylene and copolymer polypropylene waste materials. This system is very common in the industry. Then there has been a characterization of the mechanical and rheological characterization of the mixture. This characterization has allowed developing a model to predict the rheological behavior during the process of transformation; this predictive model has been experimentally validated by testing machine filling and use of specific software.

(Received March 10, 2011; accepted July 25, 2011)

Keywords: Viscosity, Mechanical recycling, Blend, Simulation

1. Introduction

In order to obtain the best trade-off between cost and final properties blend recycled polymers are usually blended with virgin polymers [1, 2].

From another point of view, the numerical simulation of the injection moulding process has acquired a great relevance for design process in product development [3].

The rheological characterization of a polymer is commonly carried out by a capillary rheometer [4]. The accuracy of numerical simulations depends on the rheological characteristics of polymers.

Our principal aim is to establish, using experimental data, which information we can correctly predict on the behavior during processing of a recycled material. We must determine which models we can use. We could carry out studies on the effect of viscosity on incompatibility of plastic waste material.

We used the mixing of two materials of the same composition or nature and which have similar viscosity [7].

2. Experimental

2.1. Materials

The first component of the system was virgin polypropylene manufactured by Repsol-YPF (PP ISPLEN® PB 180 G2M), with a MFI of 20 g/10min. The other component was recycled copolymer polypropylene from rejected components of large size which were transformed through the injection process.

2.2. Material Characterization

The mechanical properties of the samples were evaluated using an ELIB 30 electro-mechanical universal

testing machine by Ibertest (S.A.E. Ibertest, Madrid, Spain), with a load cell of 5 kN. All tests were carried out following UNE-EN ISO 527 standard, at a speed of 30 mm min⁻¹.

Melt flow index measurements were obtained with an extrusion plastometer (Ats Faar S.p.A, Vignate, Italy) equipped with a heating chamber set at 200°C where the blended granules were introduced. After a previously established melting time, the material was forced through a capillary by means of the application of a 5000 g weight.

Impact strength was determined by using the Charpy impact machine (S.A.E. Ibertest, Madrid, Spain) according to ISO-179.

The rheological characterization was carried out as described under ISO 11443 standard, using a capillary rheometer, (Rheoflizer Typ 556-0101, ThermoHaake Inc, Germany).

Tests for each sample were carried out in the three nozzles with the purpose of obtaining the real shear stress, applying the Bagley correction [8] and the real shear rate applying the Rabinowitsch correction [9].

2.3. Process and simulation

The preparation of the mixtures was carried out using a double screw extruder to ensure correct dispersion of the components in the blend [11]. To add the components to the blend, two volumetric dispensers were used.

The injection process was performed in a MATEU-SOLÉ 270/75 injection machine (MATEU-SOLÉ, Barcelona, Spain) at a screw temperature of 230°C.

Plastic flow analysis simulations were carried out using the commercial computer-aided engineering software, Autodesk Moldflow Insight 2010.

3. Results and discussion

3.1. Characterization

In the case of PP ISPLEN® PB 180 G2M,(virgin polypropylene) the mechanical properties necessary for the study were provided by the manufacturer

In the case of recycled PP table 1 shows the rheological parameters obtained from rheometer.

Table 1. Cross-WLF parameters of PP recycled

	D ₂ (K)	D ₃ (K/Pa)	\tilde{A}_2 (K)	n (-)	τ^* (Pa)	D ₁ (Pa.s)	A ₁ (-)
PP recycled	263,15	0	51,6	0,275	38004,35	2,23E+12	26,36

We have obtained a value of 12.9 g/10 min of melt index flow of the material according to ISO 1133.

According this, we acquired the real viscosity curve through a capillary rheology test (Figure 1). The testing was carried out according to ISO 11443:1995. The test temperature was set at 230°C and 240°C, as typical value for transformation of this material.

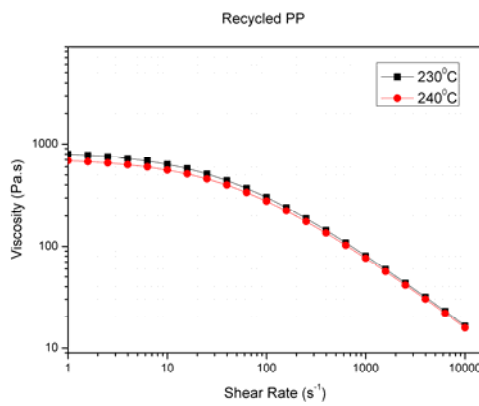


Fig. 1. Viscosity curves of recycled PP.

Finally, we obtained the mechanical properties of the material through traction and impact tests (table 2)

Table 2. Mechanical properties.

	R [MPa]	E [MPa]	ϵ_r (%)	E (kJ m ⁻²)
Recycled PP	22,2 (0,3)	532 (5)	38,7 (2)	7,78 (0,1)

3.2. Process behavior

We rheologically characterized two materials with a high compatibility index, in order to later develop a model which would allow us to evaluate the behavior of the blend during its process of transformation.

This model would later be verified experimentally using specific simulation software.

We based our study on the Cross-WLF equation and on knowledge of the physical significance of the different parameters. herefore we propose a model where, applying the blending law and we calculate the different parameters of the model for a particular mixture.

We later validated the rheological model experimentally, through mold filling tests using injection machines. For this we used a laboratory mold with normalized test samples.

These tests were carried out using the three materials characterized. Virgin Polypropylene Repsol-YPF PB 180 G2M, Recycled Polypropylene sourced from injection processes and blend of 50% by weight of both materials.

To carry out the comparative filling tests (real and simulated) we selected injection rate as the parameter under study, maintaining the rest of the parameters (screw temperature, injection time and mold temperature) constant.

We carried out filling tests, keeping the injection time constant but progressively reducing the injection rate until no flow of material was observed inside the cavity.

We obtained samples from a series of five injections from which the average weight was calculated. This operating procedure was followed in the same way for the three materials proposed.

We introduced the injection parameters used in the machine into the simulation along with the rheological characteristics of each of the materials, obtained from the Cross-WLF model parameters.

Running parallel to the filling tests in the injection machine, we carried out simulations of the process using the Moldflow® program with the aim of comparing the experimental and theoretical flow advance rates. From each of the simulations, we obtained the theoretical advance of the flow and the theoretical weight of injected material for each of the injection speeds proposed.

We saw a good correlation between the experimental flows and theoretical flows, with the exception of a small difference at low injection rate in the case of recycled material, (Fig. 2a,b,c).

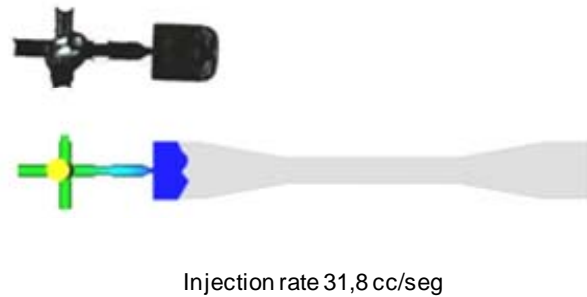


Fig. 2a. Advances in experimental and simulated flow of recycled PP at different injection rates.

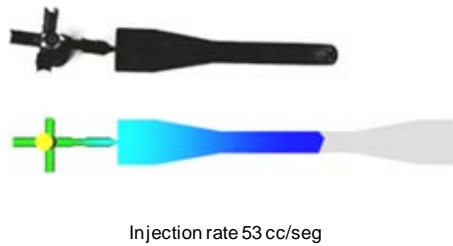


Fig. 2b. Advances in experimental and simulated flow of recycled PP at different injection rates.

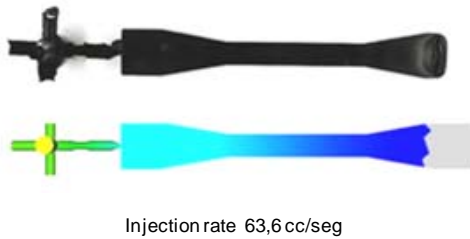


Fig. 2c. Advances in experimental and simulated flow of recycled PP at different injection rates.

3.2 Calculation of mechanical behavior

To predict behavior of polymeric blends some theoretical models have been used [12, 13]. A model, based on Takayanagi [14] has been used to evaluate quite accurately mechanical performance of blends. This model, Equivalent Box Model (EBM), considers that certain fractions of each material contribute to mechanical properties as in series work, while other fractions contribute in parallel.

The contribution of these fractions to the final value of one property is determined using the general Rule of Mixtures.

Therefore, with a 50 / 50 blend and using the critical exponent commonly used, we obtain a value for $V_{1p} = V_{2p} = 0,2$ and for $V_S = 0,6$ [15,16,18].

Composed samples of 50% each of virgin polypropylene PB 180 G2M and recycled PP, we obtained values for Young's modulus and Tensile strength which were very similar to those obtained experimentally (Table 3).

Table 3. Young's modulus (E) and Tensile strength (R) for 50/50 blend, obtained experimentally and by EMB

R (MPa)				
Virgin PP	Virgin PP	Virgin PP	Virgin PP	Virgin PP
20,5 (±0,2)	20,5 (±0,2)	20,5 (±0,2)	20,5 (±0,2)	20,5 (±0,2)
E (MPa)				
Virgin PP	Virgin PP	Virgin PP	Virgin PP	Virgin PP
495 (±1,8)	495 (±1,8)	495 (±1,8)	495 (±1,8)	495 (±1,8)

4. Conclusions

The characterization of the recycled polypropylene used in the blends shows us that no thermal degradation occurs during processing and whose behavior is very similar to that of virgin material used in the same application. The material could be considered like good quality.

The comparison between simulation and experimental results allowed us to validate the model both for the Young's modulus and Tensile strength, in which we obtained very similar values in both cases using the equivalent box model (EBM) with the parameters developed by Kolarik. This model is appropriate for the mechanical characteristics of the system.

The model proposed to calculate rheological behavior was shown to be correct as it gave rheological curves that fall between those of the two initial materials.

The rheological model proposed was shown to work accurately for the all materials studied, and we saw a good correlation between experimental and simulation flows

References

- [1] G. Lucchetta, P. F. Bariani, W.A. Knight, *Cirp Annals-Manufacturing Technology* **55**(1), 465 (2006).
- [2] R. Navarro, et al., *Journal Of Materials Processing Technology* **195**(1-3), 110 (2008).
- [3] I. Claveria, C. Javierre, and L. Ponz, *Journal of Materials Processing Technology*, **162**, 477 (2005).
- [4] P. F. Bariani, M. Salvador, G. Lucchetta, *Journal Of Materials Processing Technology* **191**(1-3), 119 (2007).
- [5] R. Scaffaro, F. P. La Mantia, *Polymer Engineering And Science*, **42**(12), 2412 (2002).
- [6] T. Boronat, et al., *Journal Of Materials Processing Technology*, **209**(5), 2735 (2009).
- [7] N. Kukaleva, G. P. Simon, E. Kosior, *Polymer Engineering And Science*, **43**(1), 26 (2003).
- [8] E.B. Bagley, *Journal Of Applied Physics*, **28**(5), 624 (1957).
- [9] B. Rabinowitsch, *Zeitschrift Fur Physikalische Chemie-Abteilung A-Chemische Thermodynamik Kinetik Elektrochemie Eigenschaftslehre*, **145**(1), 1 (1929).
- [10] A. V. Shenoy, D. R. Saini, *Rheologica Acta*, **23**(4), 368 (1984).
- [11] R. Chowdhury, M. S. Banerji, K. Shivakumar, *Journal Of Applied Polymer Science*, **104**(1), 372 (2007).
- [12] R. Hernandez, et al., *European Polymer Journal*, **36**, 1011 (2000).
- [13] J. Kolarik, *Polymer Engineering and Science*, **42**(1), 161 (2002).
- [14] M. Takayanagi, *Mem Fac Eng Kyushu Univ.* **1**, 41 (1963).
- [15] J. Quintanilla, *Polymer Engineering and Science*, **39**(3), 559 (1999).

- [16] R. Greco, M. Lavarone, *Polymer Engineering and Science*, **40**(7), 1701 (2000).
- [17] J. Kolarik, *Polymer Engineering and Science*, **40**(1), 127 (2000).
- [18] L. M. Robeson, R. A. Berner, *Journal of Polymer Science: Part B: Polymer Physics* **39**, 1093. (2001).
- [19] A. L. N. Silva, M.C.G. Rocha, F. M. B. Coutinho, *Polymer Testing*, **21**(3), 289 (2002).
- [20] M.L. Williams, R. F. Landel, J. D. Ferry, *Physical Review*, **98**(5), 1549 (1955).

*Corresponding author: sferrand@mcm.upv.es