

Effect of the Injection Moulding Processing Conditions on Biopolymers Final Properties

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Abstract

This research work intended to study the effect of the main injection moulding parameters in the final properties of biopolymers mouldings. An experimental procedure was carried out in which four biopolymers containing different composition percentages of poly-lactic acid (PLA) and plasticized starch (PLS) were compared with polypropylene (PP). For each material the effect of the processing conditions (mould temperature, injection temperature and holding pressure) on the final properties was discussed and the possibility of using biopolymers as a substitute of PP in household utility products was evaluated.

Introduction

The environmental pollution problems caused by the use of synthetic polymers based on petrochemicals, has given an extensive space and an attracted interest to the development of environmental friendly polymeric materials [1-6]. Plasticized starch (PLS) emerged as a potentially useful material for biodegradable plastics because of its natural abundance and low cost, being corn, wheat, potato and rice the main sources of starch [7]. PLS offers an interesting alternative for synthetic polymers where long-term durability is not a requirement and rapid degradation is an advantage [8-10].

For some applications, these materials have limited mechanical properties, but an increase of tensile strength can be achieved by compounding them with other biodegradable products [6,11-13]. Polylactide Acid (PLA) is one of the elected biodegradable products for this purpose, since it has good mechanical properties, easy processability and excellent degradability. So, currently, PLA is regarded as one of the most promising biodegradable polymers and is expected to substitute some of the non-biodegradable engineering plastics. However, its high cost limits its application [6,12,14].

One of the most used technologies for processing synthetic petrochemicals based polymers is injection moulding. It provides high quality polymeric parts and, if a large production is intended, it makes them available at a quite reduced cost. The aim of this paper is to study the effect of the injection moulding processing conditions on the final properties of biopolymers mouldings. To achieve this goal, an experimental protocol was set-up. Four different biopolymers were processed, with different injection conditions as regards mould temperature, injection temperature and holding pressure, and the obtained mouldings were tested. The performed tests allowed to analyse the sensitivity of the mechanical properties of each material to each of the processing parameter, and to compare the behaviour between the materials.

This work was developed with the cooperation of an SME company (FAPIL, S.A.). Its core business is currently centred in the production of a wide range of household cleaning utilities, like mops, buckets and brushes, made essentially of polypropylene (PP). The intention to launch new domestic products with a disposable nature, together with an increasing environmental

consciousness introduced the need of understanding the behavior of biopolymers and get familiar with their processing conditions.

Experimental Approach

Materials. Based on a preliminary set of material requirements, ranging from the expected life-time and normal working temperature to the need of resistance to contact with water and domestic cleaning fluids, biopolymers suppliers provided, as suitable for the application, four commercially available materials. The provided biopolymers, from three different suppliers, have a different mix content of PLA and PLS, and shown in *Table 1*, and all were used in the injection moulding experimental tests. To establish a baseline for comparison purposes, *Table 1* also presents the company most frequently used petrochemical based polymer.

Supplier Material	Code	Melt Flow Index	Melting Point	Density	Composition
Total Petrochemicals PP 9020	PP	25 [g] / 10 [min] 230°C / 2.16 kg	165 [°C]	0.905 [g/cm ³]	100% PP
Biotec Bioplast GS 2189	90/10	20-40 [g] / 10 [min] 190 °C / 2.16 kg	130 [°C]	1.2-1.4 [g/cm ³]	≈ 90% PLA ≈ 10% PLS
Cabopol Biomind R006	80/20	20-40 [g] / 10 [min] 190 °C / 2.16 kg	130 [°C]	1.2-1.3 [g/cm ³]	≈ 80% PLA ≈ 20% PLS
Rodenburg Biopolymers Solanyl 35F	40/60	~13 [g] / 10 [min] 190 °C / 2.16 kg	140-145 [°C]	1.26-1.3 [g/cm ³]	≈ 40% PLA ≈ 60% PLS
Cabopol Biomind C004	10/90	15-30 [g] / 10 [min] 100 °C / 2.16 kg	90 [°C]	1.15-1.25 [g/cm ³]	≈ 10% PLA ≈ 90% PLS

Table 1 – Material Specifications

Processing. For each parametric combination (parameter setting), 20 specimens were produced. To avoid the effect of the stabilization period of the injection process, the first 10 were rejected and only the further 10 specimens were collected for evaluation. The injection parameter setting (based on materials suppliers' information and literature review) can be seen in *Table 2*.

Test variables	Materials				
	PP	90/10	80/20	40/60	10/90
Mold temperature [°C]	30	20	20	20	20
	50	40	40	40	
Injection temperature [°C]	210	140	140	150	100
	230	160	160	170	120
	250				
Set Holding Pressure [bar]	16	30	30	30	30
	25	48	48	48	48

Table 2 – Injection parameters setting

Mechanical tests and Specimens. The evaluation of the mechanical properties was made based on tensile, flexural, impact and shrinkage tests. *Table 3* and *Figure 1* present the standards followed for testing procedures, when available, the measuring points for shrinkage evaluation and the geometry of the injected specimens.

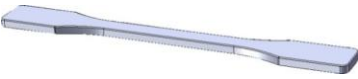

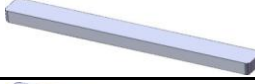

Test	Standard	Specimen
Tensile	NP-1198 (1976)	
Flexural	ASTM D790 – 03	
Impact	ISO 179:1993	
Shrinkage	-	 See Figure 1

Table 3 – Mechanical tests and Specimens

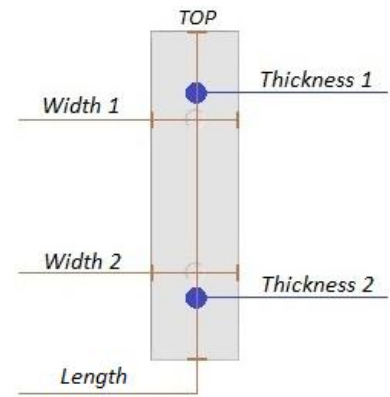


Figure 1 – Shrinkage measuring

Results

Tensile tests. Table 4 and Table 5 show the tensile tests results and the effect of the processing parameters on the results. When increasing parameter values, most materials exhibit changes in their rupture stress, as well as in the strain at rupture (Table 4). PP, 90/10 and 80/20 materials show higher resistance when subjected to tensile forces. Material 10/90 presents a very high elasticity with a strain over 130% (Table 5).

Materials	PP			90/10			80/20			40/60			10/90		
	MT	IT	HP	MT	IT	HP	MT	IT	HP	MT	IT	HP	MT	IT	HP
Maximum Stress (S_{max}) [MPa]	-	-	-	-	-	-	-	-	-	-	↓	↑		-	-
Strain (S_{max}) [%]	-	-	-	-	-	-	-	-	-	-	↓	↑		↑	↓
Rupture Stress (S_{rup}) [MPa]	↑	-	↑	↑	-	-	↑	↑	↓	↓	↓	↑		-	-
Strain (S_{rup}) [%]	↕	↕	↓	↕	↓	-	↓	↓	↑	-	↓	↑		↑	↓

MT –Mould Temperature; IT –Injection Temperature; HP – Holding Pressure
↑ – Value Increase ; ↓ – Value Decrease ; ↕ – Variable Behaviour

Table 4 – Behaviour with the increasing of the values of injection parameters (Tensile tests)

Materials	PP		90/10		80/20		40/60		10/90	
	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}
Maximum Stress (S_{max}) [MPa]	36.34	36.84	41.13	43.20	35.40	37.26	15.24	21.35	16.44	17.30
Strain (S_{max}) [%]	10.00	10.59	4.21	4.56	3.92	4.38	3.16	3.71	136.66	182.13
Rupture Stress (S_{rup}) [MPa]	20.58	25.22	17.31	20.03	15.16	28.13	14.42	20.61	15.65	16.76
Strain (S_{rup}) [%]	19.68	24.77	11.79	16.11	6.54	13.55	3.17	4.54	140.85	190.41

Val_{min} – Minimum observed value ; Val_{max} – Maximum observed value

Table 5 – Tensile test results

Flexural tests. In the Table 6 and Table 7 the flexural tests results are shown together with the effect of the processing variables on them. When increasing parameter values, most materials exhibit changes in their maximum stress (considering a strain of 1.5%), while all materials exhibit changes in the elasticity modulus (Table 6). 90/10 and 80/20 materials show the highest values of maximum stress and elasticity modulus, while the values for material 10/90 are considerably lower (Table 7 / Figure 2 and Figure 3).

Materials	PP			90/10			80/20			40/60			10/90		
Parameter	MT	IT	HP	MT	IT	HP	MT	IT	HP	MT	IT	HP	MT	IT	HP
Maximum Stress (S _{max}) [MPa] (at a strain of 1.5 %)	↑	-	-	↓	↓	-	↓	↓	↓	↓	-	-		-	-
Elasticity Modulus [MPa]	↑	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓		↓	↑

MT –Mould Temperature; IT –Injection Temperature; HP – Holding Pressure
↑ – Value Increase ; ↓ – Value Decrease ; ↓ – Variable Behaviour

Table 6 – Behaviour with the increasing of parameter value (Flexural tests)

Materials	PP		90/10		80/20		40/60		10/90	
Results	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}
Maximum Stress (S _{max}) [MPa] (at a strain of 1.5 %)	23.53	25.62	26.58	36.30	26.93	37.97	17.17	21.33	6.03	6.30
Elasticity Modulus [GPa]	1.51	1.67	2.35	2.71	2.20	2.8	1.31	1.60	0.40	0.42

Val_{min} – Minimum observed value ; Val_{max} – Maximum observed value

Table 7 – Flexural test results

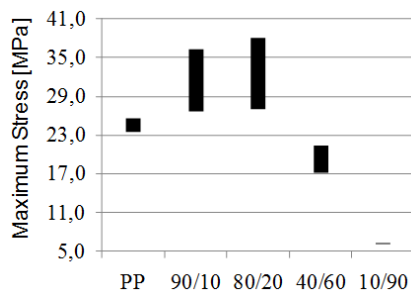


Figure 2 – Flexural tests – Maximum Stress

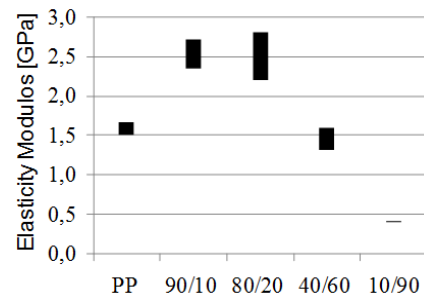


Figure 3 – Flexural tests – Elasticity Modulus

Impact tests. Table 8 and Table 9 present the results of the impact tests made on the specimens produced with different injection conditions. When processing parameters are increased, materials exhibit changes in their impact resistance (Table 8). 10/90 material shows the higher impact resistance which was 6 to 8 times higher than the one with worst results (40/60 material) (Table 9). Changes in processing parameters strongly affect the impact resistance of the materials 80/20 and 10/90, being observed deviations, between the lowest and the highest achieved values, of about 60% and 40%, respectively. However, for the same mould temperature, the sensitivity of the impact resistance to the injection temperature hardly depends on the tested holding pressures for 80/20 material (Figure 4). The same cannot be said for 10/90 material (Figure 5).

Materials	PP			90/10			80/20			40/60			10/90		
Parameter	MT	IT	HP	MT	IT	HP	MT	IT	HP	MT	IT	HP	MT	IT	HP
Impact Resistance [kJ/m ²]	↑	-	-	↑	↑	↑	↓	↓	-	↓	↓	-		↓	↓

MT –Mould Temperature; IT –Injection Temperature; HP – Holding Pressure
↑ – Value Increase ; ↓ – Value Decrease ; ↓ – Variable Behaviour

Table 8 – Behaviour with the increasing of parameter value (Impact tests)

Materials	PP		90/10		80/20		40/60		10/90	
Results	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}
Impact Resistance [kJ/m ²]	4,80	5,80	9,66	11,30	3,07	4,88	2,47	2,83	15,70	21,95

Val_{min} – Minimum observed value ; Val_{max} – Maximum observed value

Table 9 – Impact test results

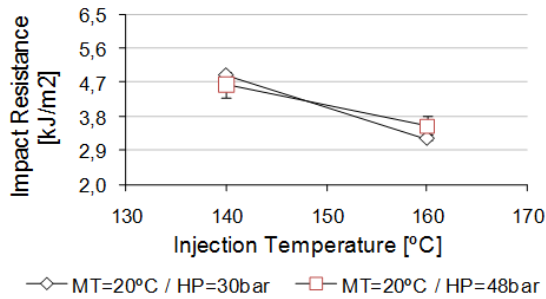


Figure 4 – Impact Resistance deviations for the material 80/20

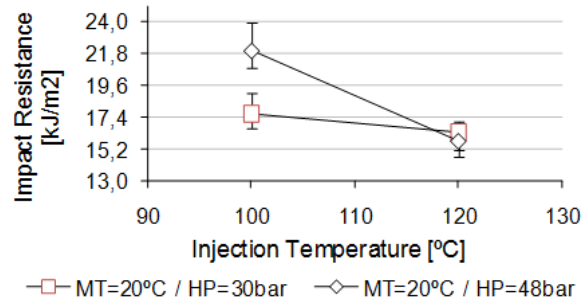


Figure 5 – Impact Resistance deviations for the material 10/90

Shrinkage tests. With different injection conditions, all materials exhibited changes in the shrinkage results (*Table 10* and *Table 11*). Specimens made of 90/10, 80/20 and 40/60 materials expanded instead of shrinking in the width direction. Moreover, the observed values of the shrinkage/expansion in width and length directions for biopolymers is much lower than for PP and were very close to zero. Between 1 and 1000 hours after processing, the observed changes were very small (*Figure 6*), being the highest shrinkage/expansion observed in the first hour after processing.

Materials	PP			90/10			80/20			40/60			10/90		
	MT	IT	HP	MT	IT	HP	MT	IT	HP	MT	IT	HP	MT	IT	HP
Width 1 [%]	↑	↑	↓	↑	↑	↓	↓	↓	↓	↓	↑	↓	↓	↑	↓
Width 2 [%]	↑	↑	↓	↑	↓	↓	↓	↓	↓	↓	↑	↓	↓	↓	↓
Length [%]	↑	↑	↓	↑	↓	↓	↓	↓	↓	↑	↑	↓	↓	↓	↓
Thickness 1 [%]	↑	↑	↓	↑	↓	↓	↓	↓	↓	↓	↓	↓	↓	↑	↓
Thickness 2 [%]	↑	↑	↓	↑	↓	↓	↓	↓	↓	↓	↓	↓	↓	↑	↓

MT –Mould Temperature; IT –Injection Temperature; HP – Holding Pressure
 ↑ – Value Increase ; ↓ – Value Decrease ; ↓ – Variable Behaviour

Table 10 – Behaviour with the increasing of parameter value (1h after processing) (Shrinkage tests)

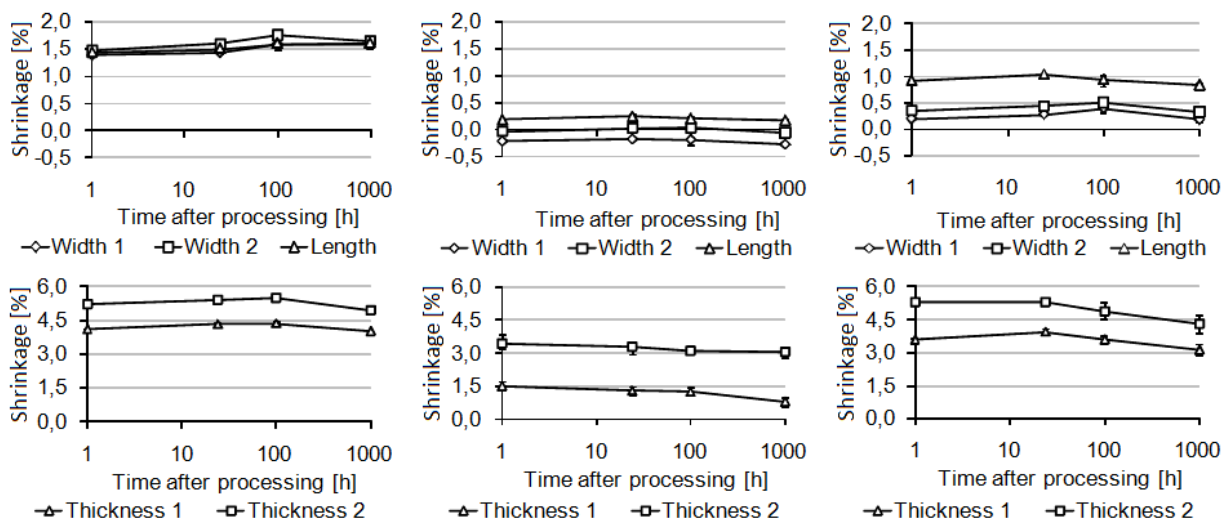


Figure 6 – Shrinkage evolution between 1h and 1000h after processing

Left > PP [MT=30°C; IT=210°C; HP=25 bar] Centre > 90/10 [MT=20°C; IT=140°C; HP=48 bar]
 Right > 10/90 [MT=20°C; IT=100°C; HP=48 bar]

Materials	PP		90/10		80/20		40/60		10/90	
	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}	Val _{min}	Val _{max}
Width 1 [%]	1.39	1.87	- 0.21	0.10	- 0.21	0.00	- 0.18	- 0.04	0.19	0.40
Width 2 [%]	1.41	1.96	- 0.03	0.12	- 0.18	0.13	- 0.13	0.12	0.18	0.47
Length [%]	1.38	1.81	0.19	0.38	0.07	0.25	0.20	0.30	0.64	0.92
Thickness 1 [%]	4.10	5.68	1.53	4.07	0.81	3.03	0.61	4.26	3.59	5.45
Thickness 2 [%]	5.13	6.81	3.44	5.80	1.21	4.38	0.38	4.77	5.31	6.84

Val_{min} – Minimum observed value ; Val_{max} – Maximum observed value

Table 11 – Shrinkage test results (1h after processing)

Conclusions

Different commercially available biopolymers with different content of PLA and PLS were tested. The achieved results show that the changes in processing conditions directly affect the final properties of the injected moulded samples. Depending on the content of PLA and PLS mechanical properties of injected biopolymers can be higher or lower than the ones observed for PP. The results point to a higher sensitivity of mechanical properties of biopolymers to changes of injection conditions as regards mould temperature, injection temperature and holding pressure. In future work, it's recommended to study the degradation of biopolymers evolution under several scenarios.

References

- [1] M. Anuradha and V.G. Kumar. Advances in contemporary research, environmentally degradable-thermoplastics: an overview. *Indian Journal Chemistry* 38 (1999) 525-531.
- [2] M. Avella, M.E. Errico, R. Rimedio and P. Sadocco. Preparation of biodegradable polyesters/high-amylose-starch composites by reactive blending and their characterization. *Journal of Applied Polymer Science* 83 (2002) 1432-1442.
- [3] L. Avérous. Biodegradable multiphase systems based on plasticized starch. *Journal of Macromolecular Science* 44(3) (2004) 231-274.
- [4] J.G. Gutierrez, P. Partal, M.G. Morales and C. Gallegos. Development of highly-transparent protein/starch-based bioplastics. *Bioresource Technology* 101 (2010) 2007–2013.
- [5] R. Narayan. Drivers for biodegradable/compostable plastics and role of composting in waste management and sustainable agriculture Report paper. *Orbit Journal* 1(1) (2001) 1-9.
- [6] Q. Shi, C. Chen, L. Gao, L. Jiao, H. Xu and W. Guo. Physical and degradation properties of binary or ternary blends composed of poly (lactic acid), thermoplastic starch and GMA grafted POE. *Polymer Degradation and Stability* 96 (2011) 175e182.
- [7] N. Cañigueral, F. Vilaseca, J.A. Méndez, J.P. López, L. Barberà, J. Puig, M.A. Pèlach and P. Mutjé. Behavior of biocomposite materials from flax strands and starch-based biopolymer. *Chemical Engineering Science* 64 (2009) 2651-2658.
- [8] M. Flieger, M. Kantorová, A. Prell, T. Rezanka and J. Votruba. Biodegradable plastics from renewable sources. *Folia Microbiologica* 48(1) (2003) 27-44.
- [9] M. Johnson, N. Tuckerb, S. Barnesc and K. Kirwan. Improvement of the impact performance of a starch based biopolymer via the incorporation of *Miscanthus giganteus* fibres. *Industrial Crops and Products* 22 (2005) 175-186.
- [10] A.K. Mohant, M. Misra and G. Hinrichsen. Biofibres, biodegradable polymers and biocomposites: an overview. *Macromolecular Materials and Engineering* 276 (3-4) (2000) 1-24.
- [11] S. Guessasma and D.H. Bassir, 2010. Identification of mechanical properties of biopolymer composites sensitive to interface effect using hybrid approach. *Mechanics of Materials* 42 (2010) 344-353.
- [12] H. Liu, F. Xie, L. Yu, L. Chen and L. Li. Thermal processing of starch-based polymers. *Progress in Polymer Science* 34 (2009) 1348-1368.
- [13] M. Wollerdorfer and H. Bader. Influence of natural fibers on the mechanical properties of biodegradable polymers. *Industrial Crops and Products* 8 (1998) 105–112.
- [14] S.L. Yang, Z.H. Wu, W. Yang and M.B. Yang. Thermal and mechanical properties of chemical crosslinked polylactide (PLA). *Polymer Testing* 27 (2008) 957–963.