

Optimization of the injection moulded PLA-cellulose composite products

T. Tábi

PhD student, Budapest University of Technology and Economics,
H-1111. Budapest, Műegyetem rkp. 3. Tel: 06-(1)-463-1459, e-mail: tabi@pt.bme.hu

F. Szabó

MSc student, Budapest University of Technology and Economics,
H-1111. Budapest, Műegyetem rkp. 3. Tel: 06-(1)-463-1459, e-mail: szabof2@indamail.hu

Abstract: *In the last few decades poly-lactic acid (PLA) became one of the most important biodegradable polymer. PLA with 5, 10 and 15weight% (wt%) cellulose content composites were produced by extrusion and 2 mm thick, 80 mm by 80 mm specimens were injection moulded. The effect of the cellulose content on the shrinkage was examined. A Taguchi-plan was also made to analyze the effect of most of the injection moulding and some mould design parameters like gate type and specimen thickness. By adding 5 or 10wt% cellulose to the PLA decreased the shrinkage, but by adding 15wt% the shrinkage increased and reached the same level as for pure PLA. By analyzing the effects of injection moulding parameters, the parameters which have the biggest effect on the shrinkage were determined.*

Keywords: *poly(lactic acid), cellulose, injection moulding, shrinkage*

1. INTRODUCTION

The importance of the plastics in the XX. century grows continuously. PlasticsEurope demonstrated that 245 million tons of plastics were processed in 2008. One third of the processed plastics are used by the packaging industry. The plastics processed for packaging typically have a short life cycle, and most of these products are only used once. Although these products are only used for a short time, their raw material is as stable as the raw material for engineering products. Furthermore the recycling of these plastics are not economical because of the low price, thus they are quickly increasing the amount of waste. As a result interest turned to bio-based and biodegradable plastics.

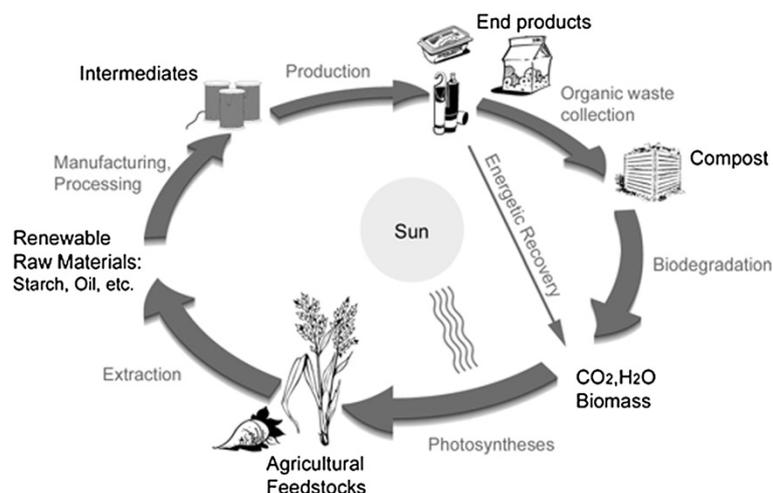


Fig.1. Life cycle of a bio-based, biodegradable polymer [1]

The full biodegradation is accomplished by the enzymatic degradation capability of bacteria, fungi and algae. During the degradation process, the molecular weight decreases, and the degraded oligomer becomes water soluble which is used by the microorganisms as a source of energy and nutrition. The end product of biodegradation are water, carbon-dioxide, biomass and inorganic compounds and in some cases methane is produced. In short: biodegradable plastics fit into the life cycle of nature (Fig. 1.) [2-5].

The Poly(Lactic Acid) (PLA) is one of the most important of biodegradable polymers. Its production increases all over the world, and extensive research is made to widen its availability and to decrease its production costs. It was not used widely until the 80', when production costs decreased significantly and recently reached a level that PLA can compete with the non-degradable petrol based plastics. PLA production is nowadays based on carbon-hydrates (starch and sugar) extracted from agricultural plants. It is possible to process PLA with extrusion, compression, injection and blow moulding. Waste bins, bottles, cutlery, mulch films can be made, but PLA can also appear as the raw material for engineering or medical products. As a drawback PLA has hydrophilic properties and is sensible for processing (heat, shearing), which can be overcome by the drying of the material and by careful processing [1, 4, 6-7].

Nowadays the price of PLA (3-4 Euro/kg) does not allow to reach wide industrial processing. In order to lower the price, increase the mechanical properties and decrease the biodegradation time, extensive research was made to blend PLA with natural products like starch or natural fibres. Most of the research is made to explore the properties of PLA blended with flax, hemp, sisal and cellulose. The usage of the fillers and reinforcing materials also have significant effect on the mechanical, shrinkage properties like in the case of petrol based plastics [8-10].

The goal of our research was to analyze the shrinkage properties of cellulose content PLA based composites, and also to examine the effect of most of the injection moulding parameters by the help of the Taguchi-method.

2. EXPERIMENTAL

5, 10 and 15wt% cellulose content PLA based composites were made using a Brabender Plasti-Corder PL 2100 type twin screw extruder (screw diameter=25 mm, L/D=20). NatureWorks Ingeo 3051D PLA and JRS Arbocel BWW40 type cellulose was used for the experiments. All of the raw materials were dried in a vented oven at 80°C for 5 hours. A melt temperature of 190°C and a screw rotational speed of 5 1/min was used. The extruded PLA/cellulose blend was pelletized, and the pellets were recrystallized at 120°C for 2 hours. 80 by 80 mm, 2 mm thick plate specimens were injection moulded with an ARBURG Allrounder 320C 600-250 (screw diameter=35 mm) type injection moulding machine. Various dimensions of the specimens were measured and used for shrinkage calculation (Fig. 2.).

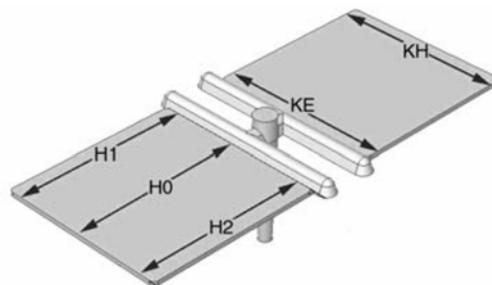


Fig.2. The places of the shrinkage measurements

The dimensions H1, H0 and H2 stand for in-flow shrinkages, while the dimensions KE and KH stand for cross-flow shrinkages. H1 and H2 are measured at the side, H0 is measured at the middle of the specimens, while KE is measured near the gate and KH is measured at the end of the flow. HSZ is introduced as the average value of the H1 and H2 shrinkages. The shrinkage measurements were made 1, 24, 48, 72 and 168 hours after injection moulding using the EN ISO 294-4:2003 standard. A Taguchi-plan (using 3 levels and 10 parameters) was made to determine the effect of the injection moulding parameters, the cellulose fibre content and some mould design parameters like specimen thickness and gate type on the shrinkage of the PLA/cellulose fibre composites. The effect of most of the main injection moulding parameters was determined. The values for the parameters were chosen as the possible lowest, highest and an average of these two. For example a holding pressure of 200-400-600 bars were analysed because warped specimens and specimens with flash could only be made using lower than 200 bars or by higher than 600 bar of holding pressure respectively.

3. RESULTS AND DISCUSSION

The shrinkage properties as the function of the cellulose content can be seen on Fig. 3.

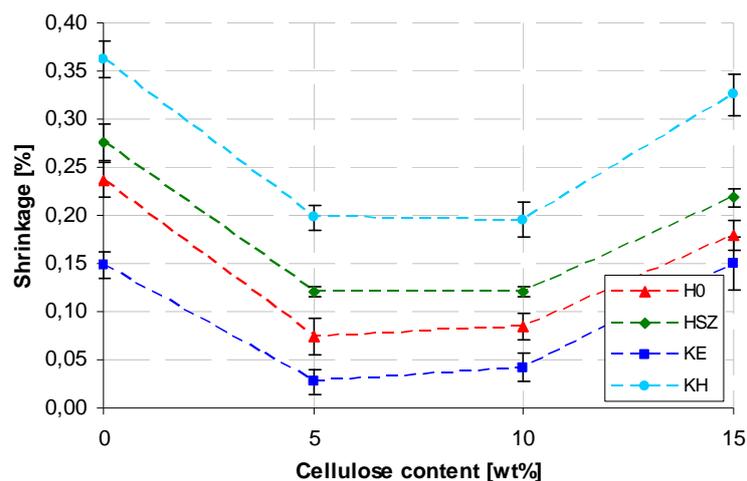


Fig.3. Shrinkage as a function of cellulose content

As it can be seen on Fig. 3. the cross-flow shrinkage measured at the gate was the lowest, while the cross-flow shrinkage measured at the end of the flow was the highest. The difference between the two cross-flow shrinkages is due to the pressure drop along the product. The shrinkage decreasing effect of the holding pressure is more effective in decreasing the cross-flow shrinkage near the gate, than at the end of the flow. The values of the in-flow shrinkages are between the two cross-flow shrinkages. The differences between the two in-flow shrinkages are mainly due to the different cooling conditions. It is found that by adding 5% and 10% cellulose to the PLA, its shrinkage decreased from an average value of 0.26% to 0.10%, while by adding 15% cellulose, the shrinkage increased to a level same as pure PLA (average value of 0.22%). The decrease in shrinkage occurred by adding 5 and 10% cellulose to the PLA is due to the shrinkage decreasing properties of natural fibres. The increasing shrinkage observed at the 10-15% cellulose fibre content region can be explained by the unusual shrinkage behaviour of pure PLA as published before [11]. The shrinkage of

the pure PLA decreases more rapidly with using more than 500-600 bars of holding pressure, than using lower holding pressures (Fig. 4.).

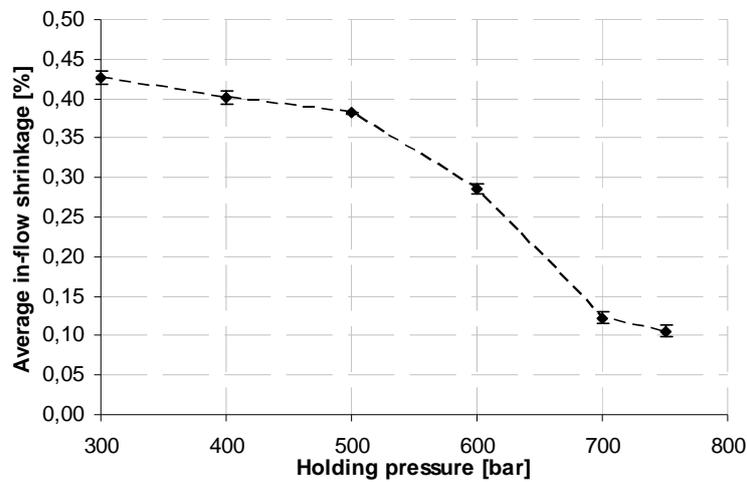


Fig.4. Average in-flow shrinkage as a function of the holding pressure for pure PLA

The viscosity of the 15% cellulose content PLA was probably higher than the viscosity of pure PLA. This is correspondent with the injection pressure need of the PLA composites, which increased from 780 to 930 (in case of 10% to 15% cellulose content) bars with adding the cellulose fibres. With the increased viscosity due to the cellulose fibres, the pressure transmitting feature of the melt decreased and the pressure inside the cavity was lower than without the fibres, thus the shrinkage increase due to the lower cavity pressure was higher than the shrinkage decrease due to the cellulose fibres. The shrinkage of the specimens was proved to be sensitive to the cavity pressure especially in the 500-700 bars holding pressure region (Fig. 4.).

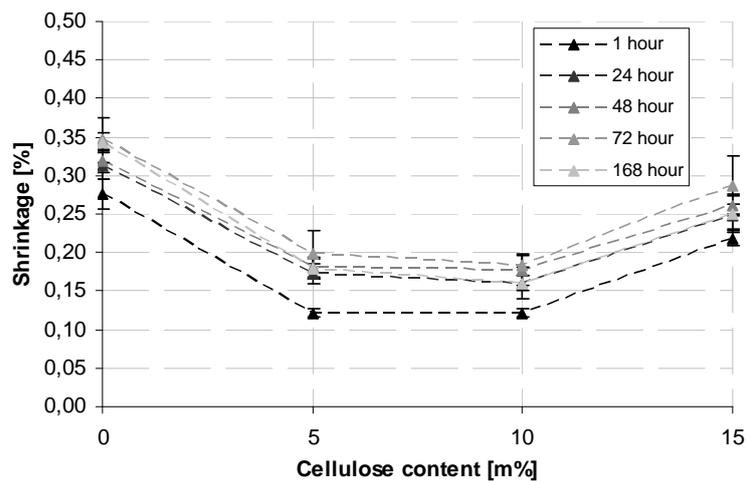


Fig.5. Shrinkage as a function of time

On Fig. 5. the shrinkage of various cellulose content PLA composites can be observed as a function of time. It is found that the shrinkage measured at different time is similar, thus with adding cellulose fibres to pure PLA has not affected the shrinkage behaviour. It is also

observed that with adding cellulose fibres, the low shrinkage of PLA can be even more lowered, which is important at mould design.

The shrinkage behaviour of injection moulded parts is affected by many independent injection moulding parameters. In order to analyse the effect of most of the injection moulding parameters on the shrinkage of PLA/cellulose composites, a Taguchi plan was made to decrease the number of measurements needed. The analysed parameters and the injection moulding set-up can be observed in Table. 1.

Table 1. The analysed injection moulding parameters

Parameter	Value
Residual cooling time	20, 30 and 40 s
Specimen thickness	1, 2 and 3 mm
Gate type	film, special film, standard
Cellulose content	5, 10 and 15%
Melt temperature	190, 200 and 210°C
Mould temperature	20, 30 and 40°C
Injection rate	40, 60 and 80 cm ³ /s
Holding pressure	200, 400 and 600 bars
Screw rotational speed	10, 15 and 20 m/min
Backpressure	30, 35 and 40 bars

The average of the cross flow and in flow shrinkages of the injection moulded specimens were used (the average of HSZ, H0, KE, KH). The measurements were made 1 hour after injecting. It was found that 4 parameters had the most effect on the shrinkage. The amplitude of the effect of different parameters can be observed in Fig. 6.

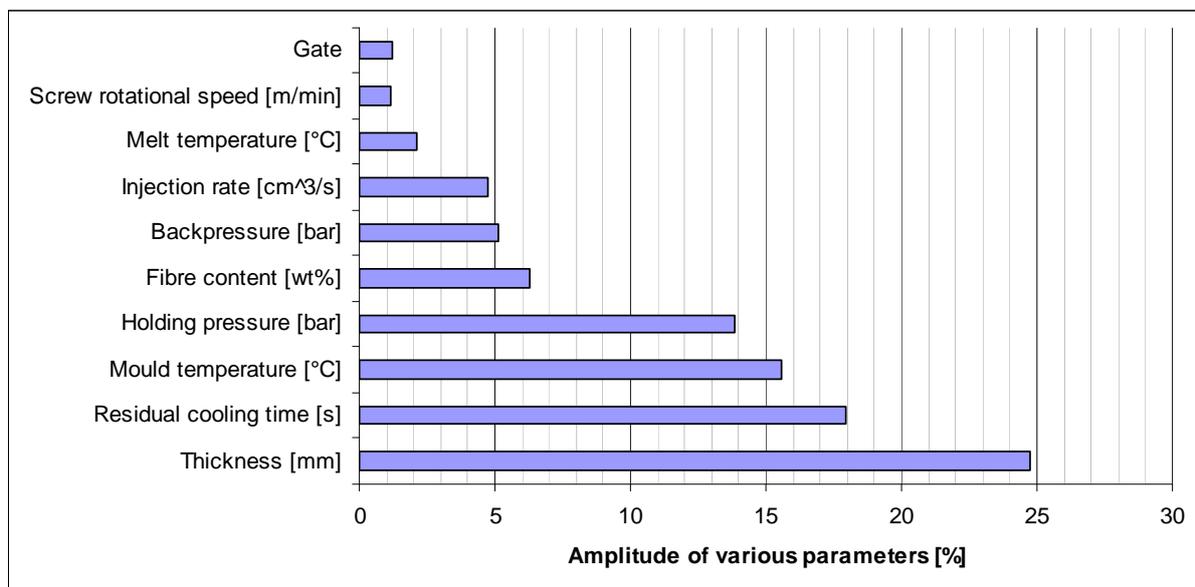


Fig.6. Amplitude of various parameters infecting shrinkage

As it can be seen, the shrinkage was mostly affected by thickness, residual cooling time, mould temperature and holding pressure. The holding pressure and the thickness also have significant effect on the shrinkage behaviour on the ordinary petrol based plastics. Similar to

ordinary petrol based plastics, the increase in part thickness and mould temperature increased, while the increase in holding pressure decreased the shrinkage.

3. CONCLUSIONS

In our work, the shrinkage behaviour of 5, 10 and 15% cellulose content PLA based composites were analysed. It was found that with adding 5 or 10% cellulose to pure PLA, its shrinkage decreased from 0.26% to 0.10%, while by adding 15% cellulose, the shrinkage increased to a level same as pure PLA (average value of 0.22%). The decreased shrinkage found at the 5 and 10% cellulose content was due to the shrinkage decrease effect of the fibres, while the increased shrinkage found at 15% cellulose content was due to higher viscosity, lower pressure transmitting feature and finally lower cavity pressure. With the help of the Taguchi-plan, the main injection moulding parameters and some mould design parameters like part thickness and gate type were investigated and found that the part thickness, the residual cooling time, the mould temperature and the holding pressure are the main parameters affecting the shrinkage.

ACKNOWLEDGEMENTS

This paper was supported by the János Bolyai Research Scholarship of the Hungarian Academy of Sciences. The authors would like to thank Arburg Hungária Kft. for the injection moulding machine, Wittmann Robottechnika Kft. for the W711 robot and the mould temperature control unit, and Lenkes GmbH. for the clamping tool system.

REFERENCES

- [1] Siracusa V., Rocculi P., Romani S., Rosa M. D.: Biodegradable polymers for food packaging: a review, *Trends in Food Science & Technology* 2008;19:634-43
- [2] The Compelling Facts About Plastics 2008, *PlasticsEurope*, Brussels, 2009
- [3] Environment Australia: Biodegradable plastics – Developments and environmental impacts, *Excelplas Australia* 2002; 1-66
- [4] Bodnár I.: Biopolimerek I.-II., oktatási segédlet, *Debreceni Egyetem, Műszaki Főiskolai Kar, Környezet-, és Vegyészmérnöki Tanszék*, Debrecen, 2005
- [5] Bastioli, C.: Handbook of biodegradable polymers, *Rapra Technology Limited*, Shawbury 2005.
- [6] Gupta A. P., Kumar V.: New emerging trends in synthetic biodegradable polymers – Polylactide: A critique, *European Polymer Journal* 2007;43:4053-74
- [7] Avérous L.: Biodegradable multiphase systems based on plasticized starch: A review, *Journal of Macromolecular Science* 2004;44:231-74
- [8] Mohanty A. K., Misra M., Hinrichsen G.: Biofibres, biodegradable polymers and biocomposites: an overview, *Macromolecular Materials and Engineering* 2000;276/277:1-24
- [9] Oksman K., Skrifvars M., Selin J.-F.: Natural fibres as a reinforcement in polylactic acid (PLA) composites, *Composites Science and Technology* 2003;63:1317-24
- [10] Iwatake A., Nogi M., Yano H.: Cellulose nanofiber-reinforced polylactic acid, *Composites Science and Technology* 2008;68:2103-06
- [11] Tábi T., Kovács J. G.: Fröccsöntött politejsav termékek vizsgálata, *Műanyag és Gumi* 2009;46:189-92