

REPROCESSING OF PLA COMPOSITES

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ABSTRACT

Biodegradable materials have been developed mainly for packaging purposes and designed for composting after use. Nevertheless, reprocessing should be also considered as an option of the waste management. In this paper influence of 10 extrusion cycles on the mechanical properties, heat deflection temperature and melt viscosity have been presented for poly(lactic acid) and PLA composites reinforced with 20% of flax fibers. It has been shown that the mechanical properties sustain the reprocessing quite well, however the melt viscosity suffers for a decrease as a result of the thermo-mechanical history developing with the subsequent extrusion cycles.

1. INTRODUCTION

Sustainable development has been considered the only viable social and economic model. The quality of life for people living in Europe depends considerably on the environment pollution. Therefore protection of the environment is one of the major challenges facing the European Union. One of its elements is the approach to waste management, which constitutes an important part of the environmental policy in the European Community. Waste management involves three complementary strategies:

- eliminating waste at source by improving product design;
- encouraging the recycling and re-use of waste;
- reducing pollution caused by waste incineration.

Every year some 2 billion ton of waste are produced in the European Community, of which only a small part constitute plastic waste. However, because of a high volume which plastic waste occupy at landfills its management is not of a marginal meaning. An important specificity of plastics is that majority of thermoplastics produced annually becomes a waste the same year. The reason is that plastics are widely used for packaging purposes, which all over the world gains 40%. Taking into account the annual consumption of plastics in Western Europe (45 Mt) one has to estimate the amount of waste plastics for 18 Mt. Such amount of the petrochemical industry products may be either landfilled, creating an accumulating ecological problem - or recycled. Fortunately, solution of ecological problems is an example of the success story at the European Community. Waste recovery and recycling has been well organized and verified within recent 10 years. Ecological legislation, recovery and recycling systems, as well as the public awareness constitute key factors of successful recycling. Packaging Directive 94/62/EC with its amendments is the fundamental act on the recovery and recycling of packaging waste at the European Community [1,2].

Recovery of the packaging waste may be performed by means of:

- mechanical recycling (reprocessing);
- chemical recycling (raw materials recovery);
- organic recycling (composting)
- incineration (energy recovery).

Majority of plastic waste is being still landfilled, but increasing quota use to be incinerated and reprocessed, in a ratio depending on a country. Incineration is a dominating option for the plastic waste management in Europe, however the amount of waste recovered by means of energy recovery cannot be counted into the obligatory recycling quota. Therefore other methods of recycling have to be developed. Mechanical recycling is developing with high dynamics, however its implementation for small objects, packaging contaminated with food waste is not economically viable. In order to meet the legislative demands, the chemical and organic recycling should be supported. Biodegradable materials have been designed for composting, as a favourable option of the waste management. However, mechanical recycling should also be considered, since while melt processing for the final shape some amounts of industrial scraps use to be generated and should be utilised, preferably by reprocessing. The objective of this study was to evaluate the influence of multiple extrusions of poly(lactic acid) (PLA) and PLA composites filled with flax fibres on their end use properties and on the melt viscosity.

2. EXPERIMENTS

2.1 Materials

Poly(lactic acid) NW2002D (NatureWorks) was used as a matrix polymer, whereas the flax fibres of 1 mm length were used as a filler (F). Polymer composites were prepared by melt mixing of PLA with flax fibres at 20 wt.% loadings using a twin screw extruder CTW 100 of the HAAKE Rheocord 9000 system at 160°C. PLA and flax fibres were dried prior to processing in order to avoid a hydrolytic degradation of poly(lactic acid). After each extrusion stage the material was grinded, then injection molded into the dumbbell samples.

2.2 Test methods

Poly(lactic acid) and PLA based composites were subjected to the uniaxial tension using the tensile machine Lloyd LR 10K. The tensile strength (TS) and Young modulus (E) were estimated using the Nexygen software.

Heat deflection temperature (HDT) was evaluated with the CEAST HDT3Vicat apparatus equipped with the oil bath.

Melt rheology was characterized with the oscillatory rheometer Haake RT20P at 160°C. Melt viscosity was measured and compared after the consecutive extrusion cycles.

3. EXPERIMENTAL RESULTS

3.1 Tensile properties

Influence of multiple extrusions on the tensile properties of poly(lactic acid) have been presented in Figure 1. The tensile strength remained almost at the same level as that after first processing. The differences within ten extrusion cycles were kept within $\pm 5\%$, which suggested a high stability of PLA towards processing.

Recycling by extrusion did not influence markedly the tensile strength of PLA/flax fibre reinforced composites, either (Figure 2). TS values did not scatter significantly during ten reprocessing cycles ($\pm 5\%$). Those findings suggest that the interactions between the flax fibres and PLA matrix remain at a similar level after several extrusions, thus transferring the tensile stress through the material for the same manner up to its catastrophic failure.

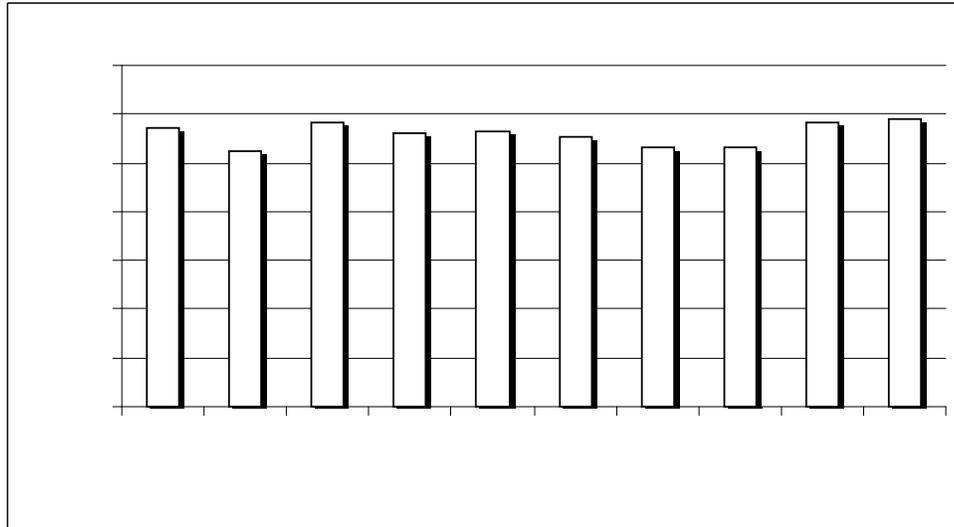


Figure 1: Tensile strength of PLA as a function of reprocessing.

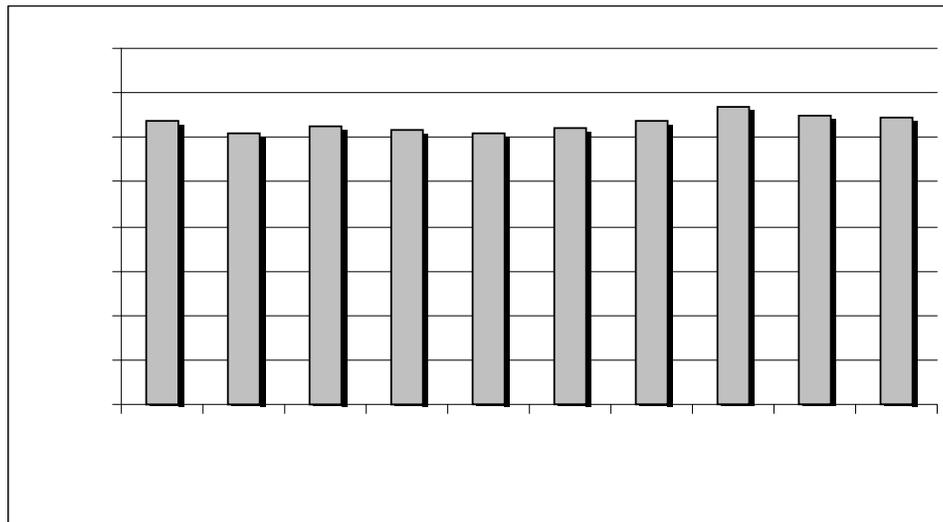


Figure 2: Tensile strength of PLA/flax fibre composites as a function of reprocessing.

Young modulus of poly(lactic acid) remained almost constant for the first five extrusion cycles (approx. 2.2 GPa), then slightly increased (Figure 3). Small differences (ca. 10%) suggest only moderate changes in the material structure, which caused its increased stiffness (Young modulus in a range of 2.4 - 2.5 GPa).

Young modulus of PLA/flax composites was markedly higher than that of poly(lactic acid), but similarly to PLA matrix, E values did not change much upon recycling, varying between 4.5 – 5.2 GPa. This finding suggests that besides the matrix polymer resistance to reprocessing, also the flax fibre/PLA interactions remain at a comparable strength level.

The results of uniaxial tensile experiments have evidenced a high resistance of PLA and PLA/flax composites to recycling. Mechanical strength of these materials is very good and allows predicting multiple use of PLA articles before they are composted after their end of life.

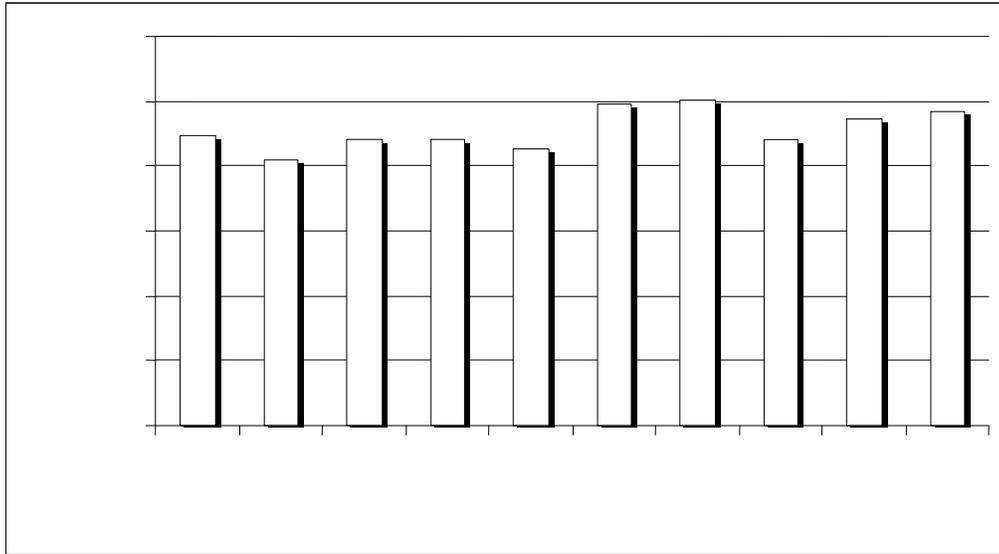


Figure 3: Young modulus of PLA as a function of recycling.

4.2 Heat deflection temperature

Heat deflection temperature exhibited also high stability during reprocessing both for PLA and PLA/flax composites, being constant within 1.5°C (Figure 4). Such behaviour confirms that also at elevated temperature the materials sustain the bending stresses without a substantial change in their structure.

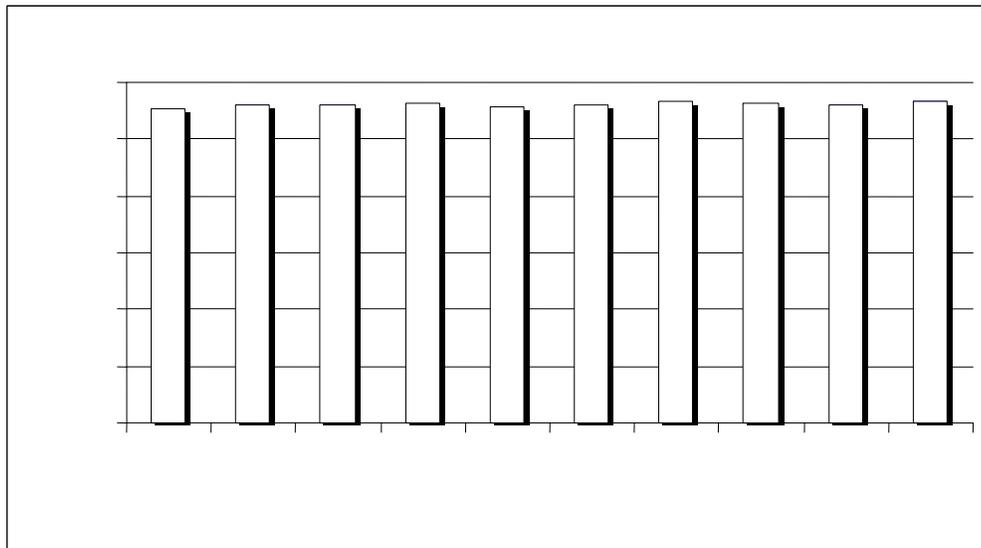


Figure 4: Heat deflection temperature of PLA as a function of extrusion cycles.

4.3 Melt viscosity

Melt rheology exhibited markedly different characteristics than that found for the materials at a glassy state. Melt viscosity of poly(lactic acid) measured at 1 rad/s decreased during first three extrusion cycles from ca. 3.9 kPa·s to around 2.5 kPa·s, thus evidencing a slight thermal degradation of the polymer (Figure 5).

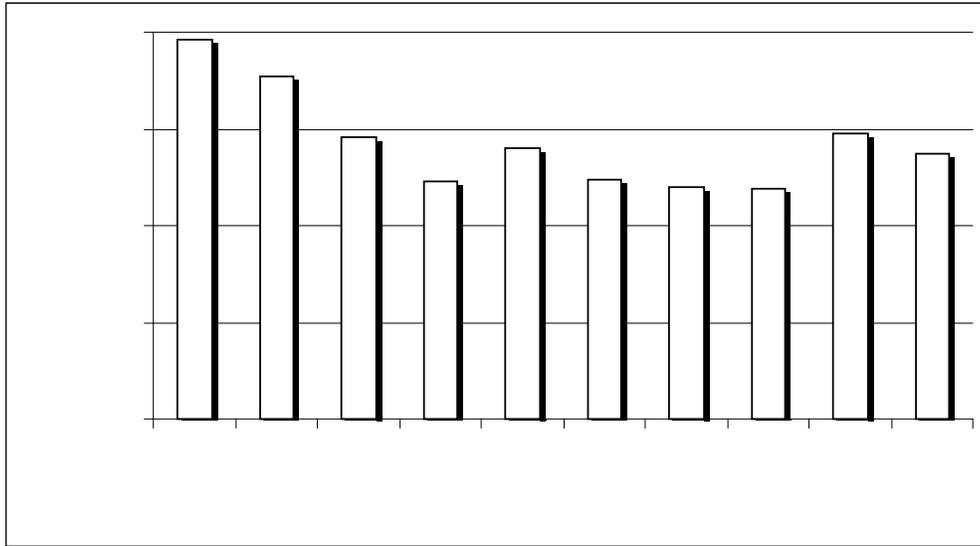


Figure 5: Melt viscosity at 1 rad/s for PLA as a function of recycling.

Rheological characteristics of PLA/flax composites was significantly different. Initial melt viscosity of the composites was almost one order of magnitude higher than that of PLA, i.e. around 30 kPa·s (Figure 6). Such behavior is regular for the fibre reinforced polymer composites. However, the consecutive reprocessing cycles brought about an exponential decrease in the melt viscosity, reaching after eight extrusions a value of 3.1 kPa·s. This reflects a highly advanced degradation of the polymer matrix, which is a totally different finding of that reported for poly(lactic acid) itself. It seems that the flax fibres caused during processing high additional stresses which, especially at small gaps in the extruder, brought about a thermo-mechanical degradation. The melt viscosity results for PLA/flax composites are radically different of the tendencies reported for the measurements at the tensile and HDT tests. This should be related to a close interrelation of the molecular weight and polymer melt viscosity, which is not a case of the mechanical properties [3].

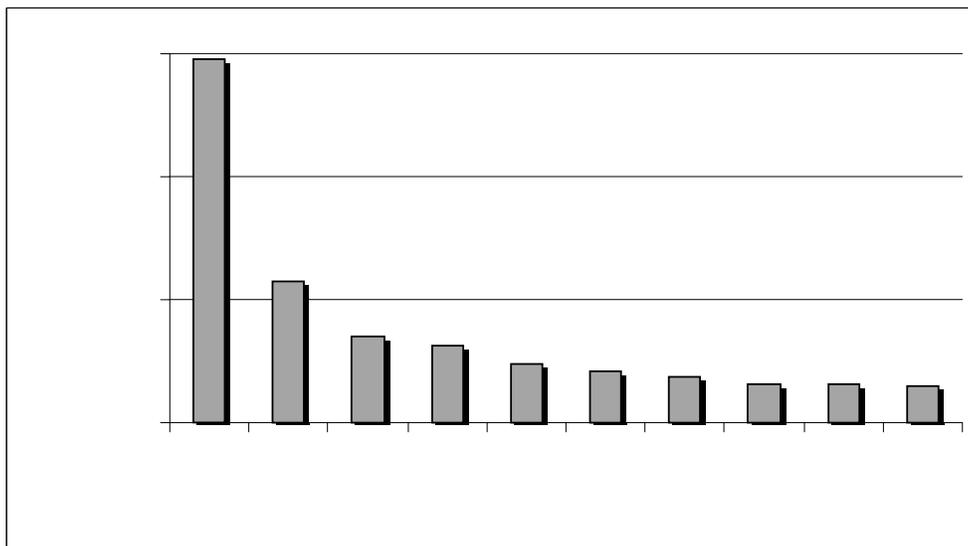


Figure 6: Melt viscosity for PLA/flax biocomposites as a function of reprocessing.

5. CONCLUSIONS

Poly(lactic acid) is a polymer of high stability during several extrusion cycles. This concerns the tensile properties, HDT and melt viscosity.

PLA/flax fibre composites exhibited high stability of the tensile properties and HDT during reprocessing. However, the melt viscosity suffered for a high decrease, thus evidencing an advanced degradation level of the polymer.

Because the mechanical properties were not markedly lower after reprocessing, the molecular weight of poly(lactic acid) did not fall below the critical value, which otherwise would cause also a dramatic decrease in the mechanical properties.

ACKNOWLEDGEMENTS

The work presented in this paper has been funded in part by the FP6 project BIOCOMP (project no. NMP2-CT-2005-515769) and by the Polish Ministry of Science and Higher Education (project no. PBZ-MNiSW-5/3/2006) under contract no G 012/T02/2007.

REFERENCES

- 1- La Mantia F.P. (Ed.), *Handbook of Plastics Recycling*, 2002, Rapra Technology, Shrewsbury.
- 2- Kozłowski M. (Ed.), *Plastics Recycling in Europe*, 2006, Oficyna Wydawnicza Politechniki Wrocławskiej, Wrocław.
- 3- Akovali G., Bernardo C.A., Leidner J., Utracki L.A., Xanthos M. (Eds.), *Frontiers in the Science and Technology of Polymer Recycling*, 1998, NATO ASI Series, vol. 351, Kluwer Academic Publ., Dordrecht