

# Effect of Natural Animal Fillers on Polymer Rheology Behaviour

M. Seidl, J. Bobek, P. Lenfeld, L. Běhálek, A. Ausperger

**Abstract**—This paper deals with the evaluation of flow properties of polymeric matrix with natural animal fillers. Technical university of Liberec cooperates on the long-term development of “green materials” that should replace conventionally used materials (especially in automotive industry). Natural fibres (of animal and plant origin) from all over the world are collected and adapted (drying, cutting etc.) for extrusion processing. Inside the extruder these natural additives are blended with polymeric (synthetic and biodegradable - PLA) matrix and created compound is subsequently cut for pellets in the wet way. These green materials with unique recipes are then studied and their mechanical, physical and processing properties are determined. The main goal of this research is to develop new ecological materials very similar to unfilled polymers. In this article the rheological behaviour of chosen natural animal fibres is introduced considering their shape and surface that were observed with use of SEM microscopy.

**Keywords**—Polypropylene matrix, Green polymers, Rheology, Natural animal fibres.

## I. INTRODUCTION

PLASTICS are world-wide spread materials. New processing technologies lead to new applications of polymers (PIM, injection molding of organic sheets etc.) which put high demands on these materials. The basic endeavour of this research is to get a tough material that should be light, easy processable and as „green“ as possible. These requirements come from needs of automotive industry where one of the biggest market with plastic parts exists. The aim is to reduce the weight of vehicles and that will lead to reduction of petrol consumption and the amount of combustion product released to air. Technical university of Liberec in cooperation with several organisations develops recipes of new polymeric materials with synthetic and biodegradable (PLA) matrix. The developed materials are given for interior parts of vehicles at this time but the final applications will be vast and will touch many industries. Branch of “green materials” is very extensive and the issue of implementation natural fibres into polymeric

matrix is very complex and that is why only the rheology is mentioned in this paper.

Deformation and flow behaviour under various conditions (different temperature and pressure) is studied by the rheology [1], [2], [3]. This branch of science works with fluid and solid mechanics. Deformation behaviour of ideal solid is described by the Hooke’s law. The applied stress is proportional to the resultant strain but is independent of the rate of this strain. When the stress is removed, the body gets the original shape again and accumulated deformation energy is released. The ideal behaviour of liquids is described by Newton’s law [4], [5]. The viscosity is defined as equivalent to the force needed to affect the flow of a fluid. Deformation behaviour of polymeric materials goes from combination of the viscosity of a liquid and the elasticity of a solid state. Properties of polymers are depend on surrounding occupation (temperature, pressure etc.) and get near to one or the other ideal state and that is why this materials are classified among viscoelastics (Fig. 1) [2], [6]. A lot of rheological models were created for description of complicated viscoelastic behaviour [7] but generally we can say that when small deformations are caused than the behaviour is liner and when bigger deformations are used than the behaviour is non-linear. Flow properties of polymeric materials can be observed in the state of melt (Non-Newtonian flow behaviour) or in the solid state (creep, stress relaxation etc.) [8]. Furthermore we will deal only with polymers in state of melt. The final melt behaviour is given by the molecular weight, chain branching, and molecular distributions and is also affected by used fillers [9].

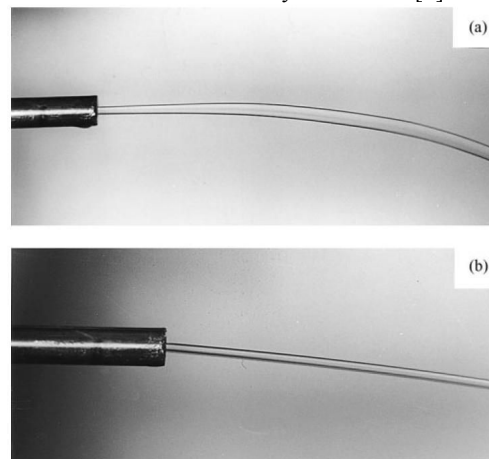


Fig. 1 Liquid jets of (a) viscoelastic fluid and (b) Newtonian fluid [2]

## II. MATERIALS AND METHODS

### A. Fillers shape and size impact

Various components are added to the polymeric matrix to reach required properties of the material. In the literature we can find two main groups of fillers. The first group involves

M. Seidl is with the Technical university of Liberec, Faculty of mechanical engineering, Plastic molding department, Liberec, 461 17, Czech republic (phone: +420485353333; fax: +420485353676; e-mail: martin.seidl1@tul.cz).

J. Bobek is with the Technical university of Liberec, Faculty of mechanical engineering, Plastic molding department, Liberec, 461 17, Czech republic (phone: +420485353164; fax: +420485353676; e-mail: jiri.bobek@tul.cz).

P. Lenfeld is with the Technical university of Liberec, Faculty of mechanical engineering, Plastic molding department, Liberec, 461 17, Czech republic (phone: +420485353350; fax: +420485353676; e-mail: petr.lenfeld@tul.cz)

L. Běhálek is with the Technical university of Liberec, Faculty of mechanical engineering, Plastic molding department, Liberec, 461 17, Czech republic (phone: +420485353331; fax: +420485353676; e-mail: lubos.behalek@tul.cz)

A. Ausperger is with the Technical university of Liberec, Faculty of mechanical engineering, Plastic molding department, Liberec, 461 17, Czech republic (phone: +420485353332; fax: +420485353676; e-mail: ales.ausperger@tul.cz)

composites materials known as “filled materials”. The volume fraction of the matrix is usually more than 50% and it is filled with some particles (talc, mica, clay etc.) [6], [10]. The shape of these particles affects all final composite properties (mechanical, rheological, physical etc.) [11]. The second group of composite materials is called “reinforced materials”. These composites are filled by short or long fibres (glass fibres, boron fibres, organic fibres, carbon fibres etc.) and volume of the matrix is usually less than 50% [6], [10]. Possible fillers orientation is shown in the Fig. 2. Each fillers formation make possible different mechanical properties [12]. What is rheology concerned, the shape, the size and the concentration of fillers have the most important effect on melt flow properties of filled materials. Viscosity increases with decreasing filler size, increasing surface area of fillers and increasing filler concentration [9].

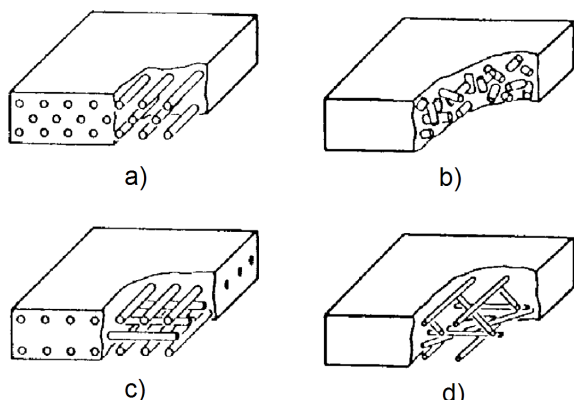


Fig. 2 Fibre orientation (a) uni-direction, (b) random, (c) orthogonal directions and (d) multiple direction [12]

#### B. Matrix impact

Fillers (especially reinforced additives) should be combined with suitable matrix for taking several advantages. The most important functions of the matrix are to keep the fillers in the structure, to protect the fillers in the structure during fabrication and also to help to transfer the load. [13]. There are a lot of different matrixes (metal, ceramic etc.) on the market but we will deal with the thermoplastics matrix. Semicrystalline thermoplastics are more efficiently reinforced than amorphous thermoplastics. In the plastic state the fillers act as nucleation sites for polymer crystallization (semicrystalline thermoplastics) and enhance the polymer crystallinity. Greater crystallinity is associated with a higher level of fibre-matrix interaction [14], [15].

#### C. Determination of melt flow index

Technological tests for determination of polymer flow behaviour are carried out on two types of instruments. Rotational viscometers are used for measurement of viscosity when small shear rate is used and capillary viscometers are more appropriate when lower viscosity with higher shear rate is measured [16]. The precise method of this test and used instruments are given by the ISO standard. Melt flow index is presented as the amount of the melt in cubic centimetre (Melt Volume Rate) or in grams (Melt Flow Rate) per 10 minutes.

For each material the test conditions are then specified (especially temperature and load). This way of determination of polymer flow properties is not universal for all polymeric materials. That is suitable only for thermoplastics whose rheological behaviour is not affected by hydrolysis or netting structure effect when increased temperatures are used [17]. The test results may predict molecular weight of thermoplastic macromolecules that directly influences the melt flow behaviour. Polymers with higher molecular weight reach better mechanical properties in solid state (higher strength, stiffness, lower tenacity) and higher viscosity [5].

### III. EXPERIMENT PROCEDURE

#### A. Material specification

For the experiment was synthetic matrix chosen. Pure polypropylene (Sumica) has very good melt flow properties and that is way the viscosity changes caused by the animal fibres will be easily proved. Polypropylene is a semicrystalline thermoplastic with relatively low strength and toughness and high tenacity. It is flammable, with no moisture absorption and it is resistant to acids, alkalis and solvents. This material is very light and can reach high level of crystallinity. Generally polypropylene with polyethylene (polyolefines) are the most world-wide used polymers [18].

As reinforced fillers hair of lama Alpaca, Camel, Angora goat (mohair) and Angora rabbit were chosen. In Fig. 3, 4, 5, 6 the pictures taken by SEM microscopy are shown. The differences among surfaces of individual fibres and their diameters are noticeable.

#### B. Compound production

Pellets of polymeric compound were produced by two screw extruder. Its construction includes two dosing areas and feeders (one for pure polypropylene matrix and the other for fibres). The screws have special design that includes two mixing zones for processing of natural fibres.

#### C. Melt flow index measurement

Determination of melt flow properties were carried out on the capillary viscometer in laboratories of Technical university of Liberec. The work area of the viscometer was heated on temperature of 180°C and the piston was weight by 2,16 kg. When the work area of viscometer was sufficiently warmed through, pellets of created compound were put into the viscometer cylinder and were compressed by the compacting rod.

Before test start the piston was put into cylinder on compressed pellets. This filling proceeding must be performed until one minute [17] and after that the test can be launched. Initially the pellets are preheated 4 minutes and then the load is put on the piston. The base principle of measurement is monitoring of piston velocity by the extensometer inside precisely given area of the cylinder. The results of the measurement are included in Table I.



Fig. 3 Picture of Angora goat fibres taken by SEM microscopy (animal picture [19])

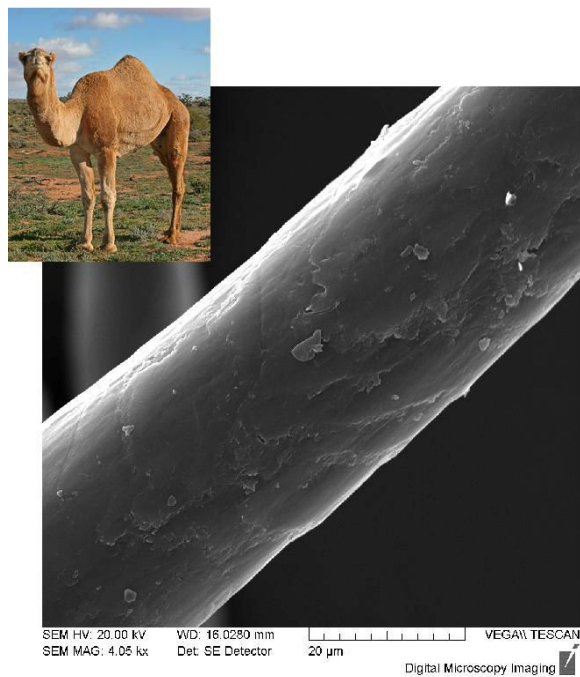


Fig. 5 Picture of Camel fibres taken by SEM microscopy (animal picture [21])

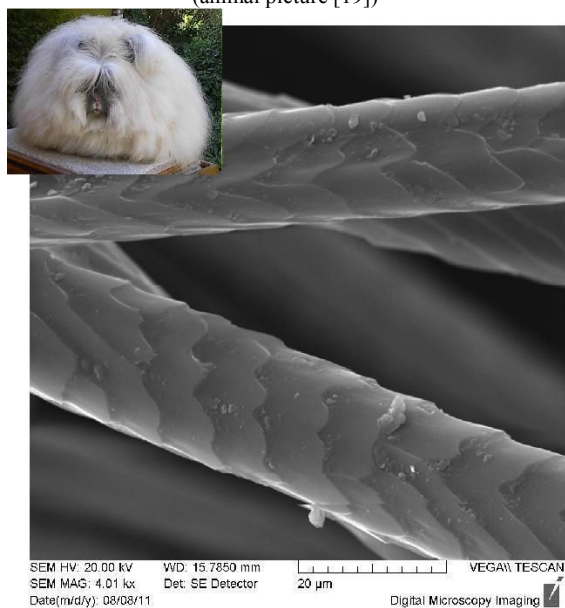


Fig. 4 Picture of Angora rabbit fibres taken by SEM microscopy (animal picture [20])

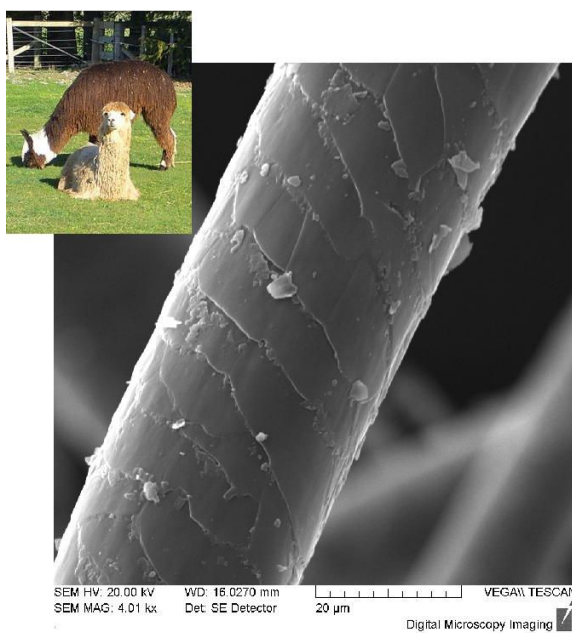


Fig. 6 Picture of lama Alpaca fibres taken by SEM microscopy (animal picture [22])

## IV. RESULTS AND DISCUSSION

TABLE I

MELT FLOW INDEX OF PP MATRIX WITH CHOSEN NATURAL ANIMAL FILLERS

Material description	ISO 1133
	MVR (180°C/2,16 kg) [cc/10 min]
PP	14,1
PP + 20 w.t.% of Angora goat fibres	6,4
PP + 20 w.t.% of Angora rabbit fibres	2,3
PP + 20 w.t.% of Camel fibres	4,1
PP + 20 w.t.% lama Alpaca fibres	1,9

The measurement results attest the theoretical foundations that reinforced fillers significantly affect the flow properties of polymer melt [9], [11], [12]. All used fibres have very similar chemical structure and are adapt in the same way (washing, drying and cutting without any other chemical modification). The length is the same for all studied fibres and their volume is 20 w.t. % in the polymeric matrix. The technological parameters for extrusion of compounded materials were also equal. All these facts mean that differences among measured melt flow indexes are caused by different shape and surface of the fillers only. Photos taken by SEM microscopy proved that fibres with the second smallest average diameter (approximately 25 µm, mohair fibres) obtain lower viscosity (higher melt volume rate) than the others. Fibres of Angora rabbit have the smallest average diameter (approximately 20 µm) but the effect of rugged surface profile is so strong that these fillers obtained one of the highest viscosity. The average diameter is identical for Camel and Alpaca fibres (approximately 30 µm). The reason of higher viscosity reached with use of Alpaca fibres is that these fillers have also more rugged surface than Camel fibres with very smooth surface profile. From the measured values the fact follows that influence of fibre surface has higher impact on the final flow behaviour than the factor of fibre size.

The research of "green and eco-friendly materials" is very beneficial and brings new materials that are suitable for a lot of applications. Our main goal is to find new formulas of polymer compounds that will have the mechanical and physical properties very similar to pure polymer matrix and to reduce the consumption of synthetic polymer in the world. Next great advantage is the price reduction of these new compounds. The concentration of 20 w.t.% natural animal fibres leads to decrease the price by approximately 16 %.

## ACKNOWLEDGMENT

This research was financially supported by Technology agency of the Czech Republic, concretely research project TA01010946. Required technical equipment was provided by Technical university of Liberec.

## REFERENCES

- [1] CARRAHER, Charles E. *Seymour/Carraher's Polymer Chemistry*. 7th edition. Florida : CRC Press, 2008. Rheology and physical tests, pp. 459-465. ISBN 978-1-4200-5102-5, ISBN 1-4200-5102-4.
- [2] HAN, Chang De. *Rheology and processing of polymeric materials : Polymer rheology*. vol. 1. Oxford : CRC Oxford university press, 2007. Relationship between polymer rheology and polymer processing, pp. 3-10. ISBN 978-0-19-518782-3.
- [3] ROSATO, Dominic V.; ROSATO, Donald V.; ROSATO, Marlene G. *Injection molding handbook*. 3rd edition. Boston : Kluwer Academic Publisher, 2000. Rheology and melt flow, pp. 530-536. ISBN 0-7923-8619-1.
- [4] CRAWFORD, Roy J. *Plastics engineering*. 3rd edition. London : Butterworth Heinemann, 2002. Analysis of polymer melts, pp. 343-346. ISBN 0-7506-3764-1.
- [5] VAN DER VEGT, A.K. *From polymers to plastics*. Delft : Delft university press/Heinemann, 2006. Viscosity, pp.103-112. ISBN 978-90-71301-62-9.
- [6] VASILIEV, Valery V.; MOROZOV, Evgeny V. *Mechanics and analysis of composite materials*. Amsterdam : Elsevier, 2001. Time and time-dependent loading effects, pp. 1,319-332,365. ISBN 0-08-042702-2.
- [7] BARNES, Howard A.; HUTTON, John F.; WALTERS, Kenneth. *An introduction to rheology*. 3rd edition. London : Elsevier, 1993. Linera viscoelasticity, pp. 37-46. ISBN 0444871403.
- [8] HUILQOL, R.R.; PHAN-THIEN, N. *Fluid Mechanics of viscoelasticity : Volume 6: General Principles, Constitutive Modelling, Analytical and Numerical Techniques (Rheology Series)*. 1st edition. Amsterdam : Elsevier, 1997. Viscometric flows, pp. 40-47. ISBN 0-444-82661-9.
- [9] HORNSBY, P. R. Rheology, Compounding and Processing of Filled Thermoplastics. In JANCA, J. *Mineral Fillers in Thermoplastics I : Raw materials and processing*. 1st edition. Berlin : Springer, 1999. pp. 155-217. ISBN 978-3-540-6421-1.
- [10] KUMAR, Anil; GUPTA, Rakesh K. *Fundamentals of polymer engineering*. 2nd edition. New York : Marcel Dekker, 2003. Step-Growth Polymerization, pp. 103-107. ISBN 0-8247-0867-9.
- [11] ROTHON, Roger N.; HANCOCK, Michael. General principles guiding selection and use of particulate materials. In ROTHON, Roger N. *Particulate-filled polymer composites*. 2nd edition. UK : Rapra technology limited, 2003. pp. 17-19. ISBN 1-85957-382-7.
- [12] CAIN, Rebecca; PINFOLD, Martyn K.; LINDSEY, Kevin A. General Properties of Composites : Stiffness, Strength and Toughness. In TUCKER, Nick; LINDSEY, Kevin. *An introduction to automotive composites*. 1st edition. UK : Rapra technology limited, 2002. pp. 59-61. ISBN 1-85957-279-0.
- [13] BERGLUND, Lars A. Polymeric matrix system : Thermoplastics resins. In PETERS, S.T. *Handbook of composites*. 2nd edition. London : Chapman & Hall, 1998. pp. 115-131. ISBN 0-412-54020-7.
- [14] CHUNG, Deborah D. L. *Carbon fiber composites*. 2nd edition. Massachusetts : Butterworth Heinemann, 1994. Polymer matrix composites, pp. 85-102. ISBN 0-7506-9169-7.
- [15] SAIELLO, S.; KENNY, J.; NICOLAIS, L. Interface morphology of carbon fibre/PEEK composites. *Journal of materials science*. 1990, 25, pp. 3496-3496. ISSN 1573-4803.
- [16] SCHRAMM, Gebhard. *A practical approach to rheology and rheometry*. 2nd edition. Germany : Thermo electro (Karlsruhe), 2004. Types of rheometers/viscosimeters, pp. 28-73. ASIN B000BWY1WA.
- [17] ISO 1133:2005. *Plastics -- Determination of the melt mass-flow rate (MFR) and the melt volume-flow rate (MVR) of thermoplastics*. Prague : Czech Standards Institute, 2005. 15 p.
- [18] BRYDSON, J. A. *Plastics materials*. 7th edition. Oxford : Butterworth Heinemann, 1999. Aliphatic polyolefins other than polyethylene and diene rubbers, pp. 247-268. ISBN 0-7506-4132-0.
- [19] *Wikipedia : The free encyclopedia* [online]. 2011-08-28 [cit. 2011-08-22]. Mohair. Accessible WWW: <<http://en.wikipedia.org/wiki/Mohair>>.
- [20] *Wikipedia : The free encyclopedia* [online]. 2011-08-21 [cit. 2011-08-22]. Angora rabbit. Accessible WWW: <[http://en.wikipedia.org/wiki/Angora\\_rabbit](http://en.wikipedia.org/wiki/Angora_rabbit)>.
- [21] *Wikipedia : The free encyclopedia* [online]. 2011-08-31 [cit. 2011-08-22]. Camel. Accessible WWW: <<http://en.wikipedia.org/wiki/Camel>>.
- [22] *Wikipedia : The free encyclopedia* [online]. 2011-06-11 [cit. 2011-08-22]. Lama Alpaca. Accessible WWW: <<http://en.wikipedia.org/wiki/Alpaca>>.