

## MELT FLOW BEHAVIOUR OF COMPOSITE MATERIALS WITH NATURAL ANIMAL FILLERS

<sup>a</sup>MARTIN SEIDL, <sup>b</sup>JIŘÍ BOBEK, <sup>c</sup>PETR LENFELD, <sup>d</sup>LUBOŠ BĚHÁLEK

Technical university of Liberec, Faculty of Mechanical Engineering, Department of Engineering Technology, Studentská 1402/2, 461 17 Liberec 1, Czech Republic  
 email: <sup>a</sup>martin.seidl@tul.cz, <sup>b</sup>jiri.bobek@tul.cz, <sup>c</sup>petr.lenfeld@tul.cz, <sup>d</sup>lubos.behalek@tul.cz

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.

**Abstract:** This paper is focused on the evaluation of flow behaviour of bio-composite materials. For this article was chosen composite with polypropylene matrix in which are natural animal fillers dispersed. The research that includes creating new materials and modifying existing ones is based on demand of using materials from renewable sources. Technical university of Liberec cooperates on long-term development of "green materials" consist of polymeric matrix and natural fibres. The basic research is focused on determination of mechanical and physical properties and processability of bio-composites and their affecting by chemical coupling agents. Except the standardised tests these materials were inspected with using of SEM microscopy to find out the influence of polymeric matrix origin, shape and size of particles and coupling agents presence on behaviour of final bio-composites.

**Keywords:** rheology, polymeric matrix, natural animal fibers, bio-composites

### 1 Introduction

Nowadays the plastic processing technologies and new approaches in chemical modification enable using of new materials. This is the best time for creating bio-composites that could replace usually used polymers. There are two ways how to achieve that. "Green materials" involves certain volume of particles of natural origin and the base (the matrix) can be of synthetics origin. The second possibility is biodegradable polymeric matrix reinforced by natural fibres that brings totally ecological composite from renewable sources. This research is basically focused on creating of new materials for automotive industry, namely for interiors parts but actually depending on used polymer matrix these materials will have very wide range of applications. Branch of "green materials" is very extensive and the issue of incorporation 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

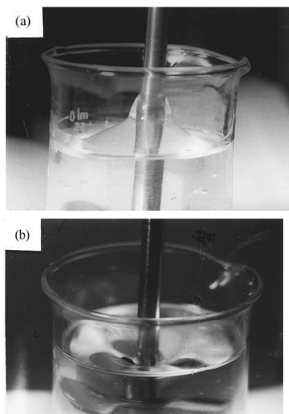


Fig.1. Different rotation behaviour between  
 (a) viscoelastic fluid and (b) Newtonian fluid [2]

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 classify 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].

### 2 Materials and methods

#### 2.1 Fillers shape and size

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 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]. Fillers can achieve different orientation depending on their shape, size and processing conditions. 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].

#### 2.2 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 and 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 thermoplasts are more efficient reinforced than amorphous thermoplasts are. In the plastic state the fillers act as nucleation sites for polymer crystallization (semicrystalline thermoplasts) and enhance the polymer crystallinity. Higher crystallinity is associated with a higher level of fibre-matrix interaction [14], [15].

#### 2.3 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].

### 3 Experiment procedure

#### 3.1 Material specification

For the experiment was synthetic matrix chosen. Pure polyacrylonitrile-butadiene-styrene has low melt flow properties due to its high molecular weight and belongs to amorphous thermoplastics. Its chemical structure makes it strength, tough with low tenacity and that is why this material is used for construction applications. Its biggest

disadvantage is quite high hygroscopic properties what is caused by polar bonds. When ABS absorbs moisture then its insulating properties are reduced.

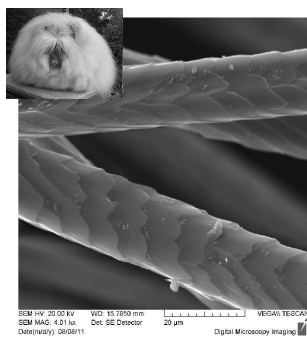


Fig.3. Picture of Angora rabbit fibres taken by SEM microscopy (animal picture [20])

#### 3.2 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.

#### 3.3 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 were compressed by the compacting rod. Before test start the piston was put into cylinder on pellets. This filling proceeding must be performed until one minute [17] and after that the test can be launched. Initially the

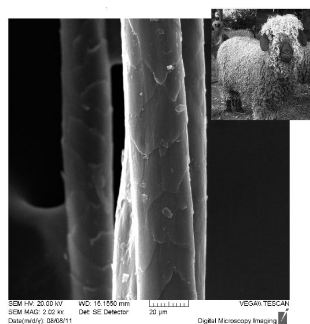


Fig.2. Picture of Angora goat fibres taken by SEM microscopy (animal picture [19])

When ABS absorbs moisture then its insulating properties are reduced. This polymer (with trade mark Magnum) is resistant to concentrate acids and alkalis [18]. As reinforced fillers hair of lama alpaca, camel, angora goat (mohair) and angora rabbit were chosen. In fig. 2, 3, 4, 5 the pictures taken by SEM microscopy are shown. The differences among surfaces of individual fibres and their diameters are noticeable.

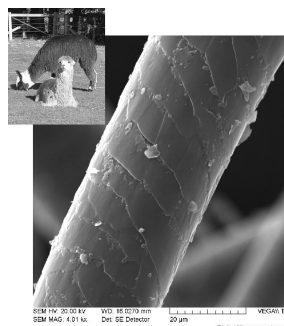


Fig.4. Picture of lama Alpaca fibres taken by SEM microscopy (animal picture [22])

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.

### 4 RESULTS AND DISCUSSION

Table I: Melt Flow Index of ABS Matrix with Chosen Natural Animal Fillers

Material description	ISO 1133
	MVR (180°C/2,16 kg) [cm <sup>3</sup> /10 min]
ABS	4,1
ABS + 20 w.t.% of Angora goat fibres	6,2
ABS + 20 w.t.% of Angora rabbit fibres	7,3
ABS + 20 w.t.% of Camel fibres	27,9
ABS + 20 w.t.% of lama Alpaca fibres	4,6

The measurement results attest the theoretical foundations that reinforced fillers significantly affect the flow properties of polymer melt [9], [11], [12]. Pure ABS has poor flow properties

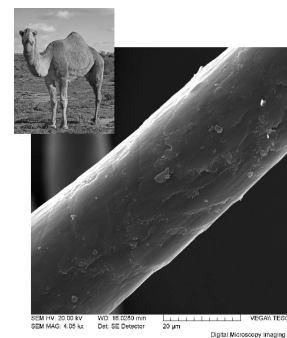


Fig.5. Picture of Camel fibres taken by SEM microscopy (animal picture [21])

at the temperature of 180 °C and all used fibres increase its melt flow index in spite of the assumption. Tested 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 20w.t. % in the polymer matrix. The technological parameters for extrusion of compound material 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 can help us to decide what has bigger impact on the melt flow behaviour if the shape profile or the size (average fibres diameter). Three types of fibres have approximately the same average diameter of 30 µm (Angora goat, Camel and lama Alpaca fibres) and so the differences of individual MVR are caused by their shape profile. The smoothest one can be seen in the Fig 5 and camel fibres also obtained the lowest viscosity (the highest melt volume rate). All the others fibres have more or less rugged surface profile. The second best result was achieved by Angora rabbit fibres because of their small average diameter. Angora goat fibres and lama Alpaca fibres look very similar and also reached similar results. They have approximately equal average diameter but the surface structure looks a little bit smoother in the case of Angora goat fibres and that is why these fibres reached better MVR. 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 reduce the consumption of synthetic polymer in the world. Next great advantage is the price reduction of these new compounds. The concentration of 20w.t. % natural animal fibres leads to decrease the price by approximately 24 %.

**Literature:**

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 JANCAR, 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. Plastics based on styrene, pp. 434-447. 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>>.

**Primary Paper Section: J****Secondary Paper Section: JI, JP**